Manifestation of New Interference Effects in Superconductor/Ferromagnet Spin Valve

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Superconductor/ferromagnet (S/F) spin valve effect theories based on the S/F proximity phenomenon assume that the superconducting transition temperature $T_c$ of F1/F2/S or F1/S/F2 trilayers for parallel magnetizations of the F1- and F2-layers ($T_c^P$) are smaller than for the antiparallel orientations ($T_c^{AP}$). Here, we report for CoO$_x$/Fe1/Cu/Fe2/In multilayers with varying Fe2-layer thickness the sign-changing oscillating behavior of the spin valve effect $\Delta T_c = T_c^{AP} - T_c^P$. We observe the full direct effect with $T_c^{AP} > T_c^P$ for Fe2-layer thickness $d_{Fe2} < 1$ nm and the full inverse ($T_c^{AP} < T_c^P$) effect for $d_{Fe2} \geq 1$ nm. Interference of Cooper pair wave functions reflected from both surfaces of the Fe2-layer appear as the most probable reason for the observed behavior of $\Delta T_c$.

More than 40 years ago Larkin and Ovchinnikov [1] and Fulde and Ferrell [2] theoretically demonstrated that a strong spin-exchange field acting on electron pairs in a superconductor may yield superconductivity with a nonuniform order parameter. In a layered superconductor/ferromagnet (S/F) thin film heterostructure such an exchange field gives rise to a damped oscillating behavior of the Cooper pair wave function which penetrates from a superconductor into a ferromagnet because of the non-zero momentum of the Cooper pairs in the F-layer. In the "dirty" limit the characteristic depth of the decay of the pairing function in the F-layer $\xi_m = (4\hbar D_m/F)^{1/2}$ is determined by the diffusion coefficient $D_m$ and the exchange splitting $I$ of the conduction band in the F-layer [3]. For pure Fe the value of $\xi_m$ is less than 1 nm (see, e.g., [4]). The oscillating behavior of the pairing function in the F-layer results in a number of new experimentally observed effects: the spatial oscillation of the electron density of states [3, 7], a nonmonotonic dependence of the superconducting (SC) transition temperature $T_c$ of F/S/F trilayers on the F-layer thickness [3, 8] and the realization of the Josephson $\pi$-junctions in S/F/S systems [3] (for a review see, e.g., [9]).

The so-called spin valve effect, a complete on/off switching of the SC current flowing in complex S/F multilayers, gives another example of the fascinating interplay between magnetism and superconductivity. Its theoretical concept developed in Ref. [10, 12] is based on the S/F proximity phenomenon and relies on the idea to control the pair breaking and correspondingly the $T_c$ by manipulating the mutual orientation of magnetizations of two layers. This is because the mean exchange field from two F-layers acting on the Cooper pairs is smaller for antiparallel (AP) orientation of magnetizations of these F-layers compared to the parallel (P) case. Theories [10, 12] predict that the AP orientation is always more favorable for superconductivity than the P one so that $T_c^{AP}$ should be always larger than $T_c^P$. However, in case of the oscillating behavior of the Cooper pair wave function in the F-layer possible interference effects in the spin valve cannot be ignored. Especially this concerns the spin valve design F1/F2/S proposed by Oh et al. [10] whose functionality should obviously critically depend on the interference at the F2/S interface of the pair wave function reflected from both surfaces of the F2-layer.

Recently we experimentally realized a full switching effect for the SC current by changing the mutual orientation of the F-layers’ magnetizations in the system CoO$_x$/Fe1/Cu/Fe2/In [13], in which the positive difference $\Delta T_c = T_c^{AP} - T_c^P$ quickly decreases from 19 to 12 mK with increasing the thickness $d_{Fe2}$ from 0.5 to 0.6 nm and finally vanishes for the sample with $d_{Fe2} = 2.6$ nm. In this system the cobalt oxide antiferromagnetic layer plays a role of the bias layer which pins the magnetization of the Fe1-layer and Cu is a normal metallic layer which decouples the magnetizations of the Fe1- and Fe2-layers.

The objective of the present work is the search of a manifestation of the quantum interference effects in an F1/F2/S spin valve system. For that we have investigated the dependence of $\Delta T_c$ on the thickness $d_{Fe2}$ of the intermediate Fe2-layer on an extensive set of spin valve samples CoO$_x$(4 nm)/Fe1(3 nm)/Cu(4 nm)/Fe2($d_{Fe2}$/20 nm) with the value of $d_{Fe2}$ lying in the range between 0.4 and 5.2 nm. We obtained clear experimental evidence for the oscillating behavior of the spin valve effect, i.e. the sign change of $\Delta T_c$ as a function of $d_{Fe2}$, which we attribute to the interference effect of the SC pairing function occurring in the F2-layer.

We used the same sample preparation method, experimental set ups and protocols of magnetic and transport measurements as in our previous work (see [13]). The residual resistivity ratio $RRR = R(300 \, K)/R(4 \, K)$ (the ratio of the electrical resistivity at 300 K to its value at 4 K) for all studied samples is similarly good evidencing a high purity of the deposited In layers. It lies in the interval $35 \leq RRR \leq 45$ corresponding to the range of the SC coherence lengths $150 \, \text{nm} \leq \xi_s \leq 170 \, \text{nm}$. For this set of samples the ratio between the In film thickness...
$d_s$ and $\xi_s$ is in the range $1.3 \leq d_s/\xi_s \leq 1.5$. This clearly shows that the dominant part of the In layer is involved into the proximity effect making the $T_c$ particularly sensitive to the ferromagnetic Fe-layers. Decreasing the In layer thickness down to $d_s \sim 180$ nm ($d_s/\xi_s \sim 1.1$) revealed that the superconductivity in this case is strongly suppressed yielding $T_c$ below 1.4 K.

Experimentally we focussed on the determination of both the hysteresis magnetization behavior and the current in-plane transport measurements, enabling a correlation between both properties. To determine the magnetic field range where AP and P states can be achieved the in-plane magnetic hysteresis loops of samples in the direction of the magnetic field along the easy axis were measured by a SQUID magnetometer. Fig. 1 shows the magnetic hysteresis loop for the sample with $d_{Fe2}=1.3$ nm. The shape of this loop is similar for all studied samples. In order to pin the magnetization of the Fe1-layer by the bias CoO$_2$-layer the sample was cooled down in a magnetic field of +4 kOe applied parallel to the sample plane and measured at $T=4$ K. As a result in the field range between -0.3 and -3.5 kOe the mutual orientation of two F-layers is antiparallel because the magnetization of the Fe1-layer is kept by the bias CoO$_2$-layer.

A minor hysteresis loop was obtained by measuring the magnetization $M(H)$ with decreasing the field from +4 kOe down to -1 kOe and increasing the field again up to +1 kOe. These minor loops for two representative samples with $d_{Fe2}=0.5$ and 1.3 nm are shown in Figs. 2a,b. As expected, the magnitude of the change of $M$ is proportional to the thickness of the free F2-layer. The coercive field $H_c$, which is of the order of 30 Oe for samples with $d_{Fe2} <1$ nm, appreciably decreases down to $H_c \leq 20$ Oe with increasing the $d_{Fe2}$ above 1 nm. This clearly shows that the domain state in the F-layers is progressively confined to smaller fields with increasing the $d_{Fe2}$. Usually in thin films the Neel type domain structure is realized. A decrease of $H_c$ with increasing the Fe2-layer thickness takes place due to a decrease of the pinning of the domain walls by interfaces arising due to the roughness and surface anisotropy.

In order to study the influence of the mutual orientation of the magnetizations on $T_c$ we have recorded the change of the resistivity upon transition to the SC state at the two in-plane switching field values of $+H_0$ and $-H_0$. First, the samples were cooled down from room temperature to $T=4$ K at a magnetic field of 4kOe applied along the easy axis of the sample just as we did it when performing the magnetization measurements. For this field both F-layers’ magnetizations are aligned. Next, the $T$-dependence of the resistivity $R(T)$ was recorded at 7 fixed values of the in-plane field $\pm H_0$ in the range $|H_0|=0-110$ Oe (examples see in Fig. 3). The dependencies of the width of the SC transition $\delta T_c$ and the magnitude of the spin valve effect $\Delta T^*_c = T_c(-H_0) - T_c(H_0)$ on $H_0$ are shown in Figs. 2c-2d and 2e-2f, respectively, for two samples with $d_{Fe2}=0.5$ nm and 1.3 nm where they can be compared with the respective minor hysteresis loops (Fig.2a, b). Both quantities have a clear correlation with the occurrence of the domain state: The presence of magnetic domains broadens the SC transition and the spin valve effect can only be observed when domains are suppressed by magnetic field, i.e. when the AP and P states are achieved. In this limit $\Delta T^*_c$ becomes equal to $\Delta T_c$. Remarkably, the sign of $\Delta T_c$ is positive for $d_{Fe2} <1$ nm and is negative for $d_{Fe2} \geq 1$ nm.

The detailed dependence of the spin valve effect $\Delta T_c$ on the thickness of the F2-layer is shown in Fig. 4. $\Delta T_c$ first increases with increasing $d_{Fe2}$ and has a sharp maximum of 19 mK at $d_{Fe2}=0.5$ nm. For larger thicknesses $\Delta T_c$ decreases down to a very small value of 4 mK for
The spin valve effect based on the S/F proximity effect predicts only the direct effect, i.e., $\Delta T_c > 0$. The inverse effect with $T_{cAP} < T_{cF}$ has been reported earlier for various systems. It’s 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. 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 $T_c$. Another mechanism also involving domains invokes an interaction between outer F-layers in the F/S/F trilayer, e.g., due to stray-field-induced magnetoelastic coupling. It yields the inverse effect since the suppression of domains in the fully polarized state with the parallel alignment of the magnetizations of the F-layers drastically reduces the domain induced stray fields normal to the layers which otherwise act on the S-layer and suppress the superconductivity.

Model (ii) is based on the giant magnetoresistance effect and predicts an enhanced spin-dependent reflection of the spin-polarized charge carriers 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. Its origin was discussed in two different ways. Rusanov et al. 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 $T_c$. Another mechanism also involving domains invokes an interaction between outer F-layers in the F/S/F trilayer, e.g., due to stray-field-induced magnetoelastic coupling. It yields the inverse effect since the suppression of domains in the fully polarized state with the parallel alignment of the magnetizations of the F-layers drastically reduces the domain induced stray fields normal to the layers which otherwise act on the S-layer and suppress the superconductivity.

For the interpretation of the negative sign of $\Delta T_c$ for thicknesses $d_{Fe2} \geq 1$ nm scenario (i) should be considered seriously since even if the exchange coupling between the F-layers is negligible due to a thick enough 4 nm Cu-interlayer, the purely magnetostatic reason for domain formation could play a role. A magnetic stray field normal to the In layer, which is a type I superconductor, might lead then to a direct suppression of superconductivity. Indeed, the presence of the domain structure may be responsible for the broadening of the SC transition at small fields due to inhomogeneous magnetic field produced by the domain walls (Fig. 2c). However, for the values of the switching field $H = \pm 110$ Oe used to determine the $\Delta T_c(d_{Fe2})$-dependence in Fig. 4 the magnetization of F1- and F2-layers are already saturated in the P or AP configurations (Fig. 2b). The incomplete saturation where the domain induced stray fields normal to the S-layer may arise is restricted to a small range of magnetic fields $-50$ Oe $< H_0 < 50$ Oe for the samples with $d_{Fe2} \geq 1$ nm, but just in this field range the inverse spin valve effect is smallest compared to the values measured outside this range (Fig. 2f). Therefore we conclude that the influence of the domain structure on the observed spin valve effect is negligibly small in our samples.

Scenario (ii) based on the spin imbalance mechanism can be surely excluded in our case because the enhanced spin-dependent reflection of spin-polarized carriers at the S/F interfaces in the AP state of the F/S/F multilayer.

![Figure 3](image-url) (Color online) The resistive superconducting transition curves for the samples with $d_{Fe2}=0.5$ nm (a - $H=110$ Oe, c - $H=20$ Oe) and 1.3 nm (b - $H=110$ Oe, d - $H=20$ Oe).

![Figure 4](image-url) (Color online) The dependence of the $T_c$ shift $\Delta T_c = T_{cAP} - T_{cF}^0$ on the Fe2-layer thickness $d_{Fe2}$. The applied switching field $H = \pm 110$ Oe. Theoretical curve (solid line) corresponds to the calculated function $[W(0)-W(\pi)]/W(0)$ (see Fominov et al. (22)) normalized to our experimental data. This fit gives $\xi_m = 0.9$ nm, $t_m = 1.5$ nm and a rough estimate of the quantum mechanical transparency of the S/F interface for the electrons $T = 0.7$ (see the text).

d_{Fe2} = 0.8$ nm suggesting a complete vanishing of the spin valve effect at even larger thicknesses. Surprisingly, further increasing the $d_{Fe2}$ in the interval $1 \text{nm} \leq d_{Fe2} \leq 2.6$ nm reveals the recovery of the effect but with the negative sign. $\Delta T_c(d_{Fe2})$ reaches its maximum negative value of $-33$ mK for $d_{Fe2} = 1.2$ nm and above this thickness smoothly approaches zero. Thus, the $\Delta T_c(d_{Fe2})$-dependence exhibits a remarkable oscillating behavior in the thickness range $0.5 \text{nm} \leq d_{Fe2} \leq 2.6$ nm.

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. The theory of
trilayer but not in our F1/F2/S structures where the S-layer is not sandwiched between the F-layers.

As to scenario (iii), indeed in a ferromagnetic layer the Cooper pair acquires a non zero momentum due to the Zeeman splitting of electron levels and thus its wave function should oscillate in space (see, e.g., [3]). 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. 4).

Interestingly, there is a recent theory developed by Fominov et al. [23, 24] 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 \( (l_m < \xi_m) \) were assumed. Here \( l_m \) is the mean free path of conduction electrons. In our samples the F-layer made of iron is a strong ferromagnet with \( \xi_m \sim 1 \) 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 [3]. Also the “dirty” limit is not realized owing to a small value of \( \xi_m \). 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 \( \Delta T_c (d_{F2}) \)-dependence in Fig. 4. An appreciable discrepancy with the experimental data point \( d_{F2} = 0.4 \) nm occurs most probably because at this thickness a transition from a continuous to an island like Fe film at even smaller thicknesses \( d_{F2} \) does take place. The fit parameters turned out to be quite realistic. We obtained \( \xi_m = 0.9 \) nm and \( l_m = 1.5 \) nm, confirming that our samples satisfy the “clean” limit \( (l_m > \xi_m) \), and the quantum mechanical transparency of the S/F interface for the electrons \( T = 0.7 \). The reasonable values of the fit parameters and the fact that the theory correctly describes the observed oscillation of the \( T_c \)-shift \( \Delta T_c \) gives additional arguments favoring the S/F proximity effect as the origin of our striking observation, which however stands on its own regardless a specific theoretical model.

In summary, we have 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 \( \xi_m \) of the Cooper pair wave function in the F2-layer and the inverse spin valve effect for larger thickness up to 2.5\( \xi_m \). 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.

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