Peculiarities of performance of the spin valve for the superconducting current

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The spin valve effect for the superconducting current based on the superconductor/ferromagnet proximity effect has been studied for a CoO_x/Fe1/Cu/Fe2/Cu/Pb multilayer. The magnitude of the effect $\Delta T_c = T_c^{AP} - T_c^{P}$, where T_c^{P} and T_c^{AP} are the superconducting transition temperatures for the parallel (P) and antiparallel (AP) orientation of magnetizations, respectively, has been measured for different thicknesses of the Fe1 layer d_{Fe1}. The obtained dependence of the effect on d_{Fe1} reveals that ΔT_c can be increased in comparison with the case of a half-infinite Fe1 layer considered by the previous theory. A maximum of the spin valve effect occurs at $d_{Fe1} \sim d_{Fe2}$. At the optimal value of d_{Fe1} almost full switching from the normal to the superconducting state when changing the mutual orientation of magnetizations of the iron layers Fe1 and Fe2 from P to AP is demonstrated.

The possibility to create a spin valve, based on the superconductor/ferromagnet (S/F) proximity effect is actively studied both theoretically and experimentally. Two different constructions of the spin valve for the superconducting current have been theoretically proposed. The first one [1] is the F1/F2/S multilayer system where F1 and F2 are the ferromagnetic layers with uncoupled magnetizations, and S is the superconducting layer. Calculations [1] show that at parallel (P) orientation of magnetizations of F1 and F2 layers the superconducting transition temperature T_c (T_c^{P}) is lower than in the case of their antiparallel (AP) orientation (T_c^{AP}). The second construction [2, 3] is F1/S/F2. Its operation is similar to the first one. Several experimental works confirmed the predicted effect of the mutual orientation of the magnetizations in the F/S/F structure on T_c (see, e.g., [4–9]). However, the magnitude of the spin valve effect $\Delta T_c = T_c^{AP} - T_c^{P}$ turned 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. Constructions similar to that suggested in [1] were studied to a

less extent [10–12]. Theoretical works by Fominov et al. [13, 14] have generalized the theory of the spin valve effect for both constructions taking into account the appearance of the triplet component in the superconducting condensate. Recently, when studying the construction proposed by Oh et al. [1] on an example of multilayer CoO_x/Fe1/Cu/Fe2/In, we have succeeded to obtain a full switching between the superconducting and the normal state when changing the mutual orientation of the magnetizations of F1 and F2 layers [15]. (Here CoO_x is an antiferromagnetic bias layer which fixes the magnetization of the Fe1 layer along the cooling field direction, Cu is a nonmagnetic layer N which decouples the magnetizations of the Fe1 and Fe2 layers, and In is a superconducting indium layer.) Furthermore, a detailed study of the spin valve effect has shown that the magnitude of the effect ΔT_c strongly depends on the Fe2 thickness d_{Fe2} yielding the change of its sign at large values of d_{Fe2} [16, 17].

To improve the operating parameters of the system (in particular, to increase T_c) we have replaced In by Pb [18] and have found out that the full switching can not be achieved because of a large value of the superconducting transition width δT_c. It should be noted that the

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thickness of the Fe1 layer should also affect the value of ΔT_c. This is because the mean exchange field from two F-layers acting on Cooper pairs in the space between Fe1 and Fe2 layers should be compensated for the AP orientation of the magnetizations of Fe1 and Fe2 layers. Thereby a naive consideration shows that for the observation of the maximal spin valve effect it is desirable to have comparable values of d_{Fe1} and d_{Fe2}.

The basis of the present work has been formed by our earlier studies of the superconducting spin valve effect in the multilayer system $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Cu}/\text{Pb}$ [18]. In the present paper we study a dependence of ΔT_c on the thicknesses of the Fe1 layer d_{Fe1} and of the Fe2 layer d_{Fe2}. At the optimal value of d_{Fe1} we have succeeded to demonstrate an almost full switching from the normal to the superconducting state.

The layer sequence $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Cu}/\text{Pb}$ was deposited on a single-crystalline MgO substrate (Fig. 1). We used the same sample preparation method, experi-
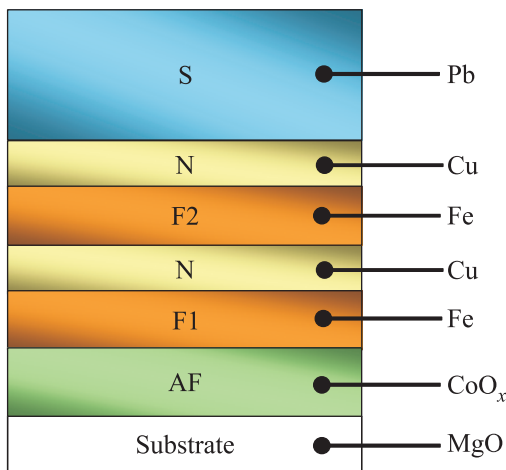

Fig. 1. Design of the studied samples

mental setups, and protocols of magnetic and transport measurements as in our previous work [15]. An additional copper layer between Fe and Pb layers was evaporated in order to stabilize the properties of the Fe2/Pb interface. Full characterization of these samples shows that this additional layer does not affect the spin valve effect. At the final step of the preparation process all samples were capped by a 85 nm thick SiN dielectric layer to protect the structure from oxidation.

For all samples we have performed magnetization measurements using a VSM SQUID (vibrating sample magnetometer superconducting quantum interference device) magnetometer. We have measured the major and minor hysteresis loops $M(H)$ in order to determine the field range in which the full switching between

P and AP orientations of the magnetizations of the Fe1 and Fe2 layers is realized. We have found out that the hysteresis loop related to the free iron layer Fe2 saturates at a field of the order of ± 1 kOe suggesting a complete suppression of the domain state.

Electrical resistivity measurements were performed with a standard four-point probe setup in the dc-mode. We have combined the electrical setup with a high homogeneous vector electromagnet that enables a continuous rotation of the magnetic field in the plane of the sample and have used a system which enables a very accurate control of the real magnetic field acting on the sample. The magnetic field strength was measured with an accuracy of ± 0.3 Oe using a Hall probe. The temperature of the sample was monitored by the $230\,\Omega$ Allen–Bradley resister thermometer which is particularly sensitive in the temperature range of interest. Therefore the accuracy of the temperature control within the same measurement cycle below 2 K was better than $\pm 2-3$ mK. To avoid the occurrence of the unwanted out-of-plane component of the external field the sample plane position was always adjusted with an accuracy better than $3°$ relative to the direction of the dc external field.

In order to study the influence of the mutual orientation of the magnetizations on T_c we have cooled down the samples from room to a low temperature at a magnetic field of 4 kOe applied along the easy axis of the sample just as we did it when performing the SQUID magnetization measurements. For this field both the F-layers' magnetizations are aligned. Then at the in-plane magnetic field value of $\pm H_0 = \pm 1$ kOe the temperature dependence of the resistivity R was recorded.

Fig. 2 depicts the dependence of the magnitude of the spin valve effect $\Delta T_c = T_c^{\text{AP}} - T_c^{\text{P}}$ on the thickness of the Fe2 layer for fixed thickness of the Fe1 layer $d_{\text{Fe1}} = 2.5$ nm[2]. For samples with $d_{\text{Fe2}} < 0.95$ nm we have observed the direct effect with $T_c^{\text{P}} < T_c^{\text{AP}}$, whereas for samples with $d_{\text{Fe2}} > 0.95$ nm the inverse effect with $T_c^{\text{P}} > T_c^{\text{AP}}$ has been found. The data shown in Fig. 2 are qualitatively rather similar to our previous results on the sign changing oscillating spin valve effect in $\text{CoO}/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{In}$ multilayers [15–17]. Practically, in our samples the iron layers thinner than 0.5 nm are not continuous anymore. The reason why δT_c for the Pb layer in contact with the Fe layer is larger than that for the In layer in contact with the same Fe layer is unclear.

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[2] Experimental points on this figure are taken from our previous paper [18]. However, the theoretical curve is slightly changed in order to describe simultaneously the experimental results shown in Figs. 2 and 3.
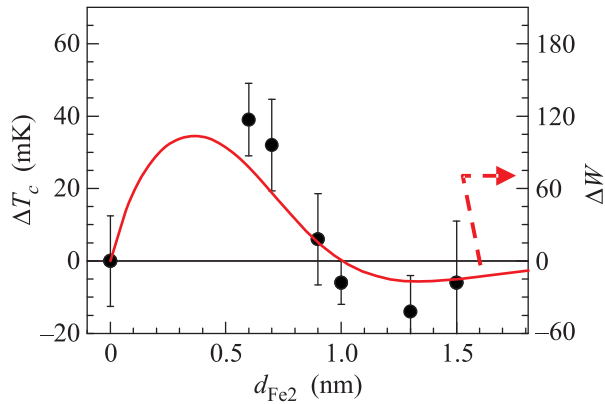
Fig. 2. Dependence of the magnitude of the spin valve effect ΔT_c on the thickness of the Fe2 layer at a fixed values of the S layer $d_{\mathrm{Pb}} = 35\,\mathrm{nm}$ and Fe1 layer $d_{\mathrm{Fe1}} = 2.5\,\mathrm{nm}$. Solid line is a theoretical curve for ΔW (see the text)

Fig. 3 shows the dependence of the magnitude of the spin valve effect $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$ on the thickness of
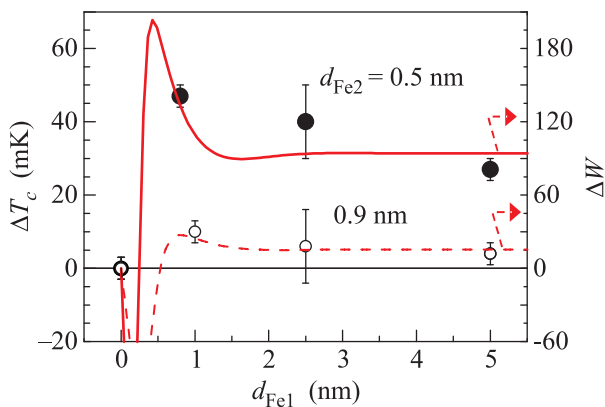

Fig. 3. The dependence of the T_c shift $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$ on the Fe1 layer thickness d_{Fe1} for the series of the samples with $d_{\mathrm{Fe2}} = 0.5\,\mathrm{nm}$ (•) and $0.9\,\mathrm{nm}$ (○) at fixed $d_{\mathrm{Pb}} = 35\,\mathrm{nm}$. The applied switching field $H_0 = \pm 1$ kOe lies in the plane of the film. Solid and dashed lines are theoretical curves for ΔW (see the text)

the Fe1 layer for two fixed thicknesses of the Fe2 layer. One can see that the dependences of $\Delta T_c(d_{\mathrm{Fe1}})$ have maximum at the values of d_{Fe1} of the order of 1 nm or less. The maximum in $\Delta T_c(d_{\mathrm{Fe1}})$ can be related to the compensation of the mean exchange field in the space between Fe1 and Fe2 layers occurring for nearly equal thicknesses of these two layers. With increasing the d_{Fe2} value the spin valve effect diminishes. This is because the penetration depth of the Cooper pairs into the iron layer is $\xi_{\mathrm{F}} \simeq 0.8\,\mathrm{nm}$ [19]. That means that only a small amount of the Cooper pairs can penetrate through the

Fe2 layer to be subjected to the influence of the Fe1 layer.

For the spin valve sample $CoO_x/Fe1(0.8$ nm$)/Cu(4$ nm$)/Fe2(0.5$ nm$)/Cu(1.2$ nm$)/Pb(60$ nm$)$ the difference in T_c for different magnetic field directions is clearly seen (see Fig. 4). The superconducting transition temperature for the AP orientation of the magnetizations occurs at a temperature exceeding T_c for the P orientation of the sample by 40 mK, which is of the order of the superconducting transition width δT_c. This opens a possibility to switch off and on the superconducting current flowing through our samples *almost 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 resistivity measurements of the sample by sweeping slowly the temperature within the ΔT_c and switching the magnetic field between $+1$ and $-1\,\mathrm{kOe}$ (Fig. 4).
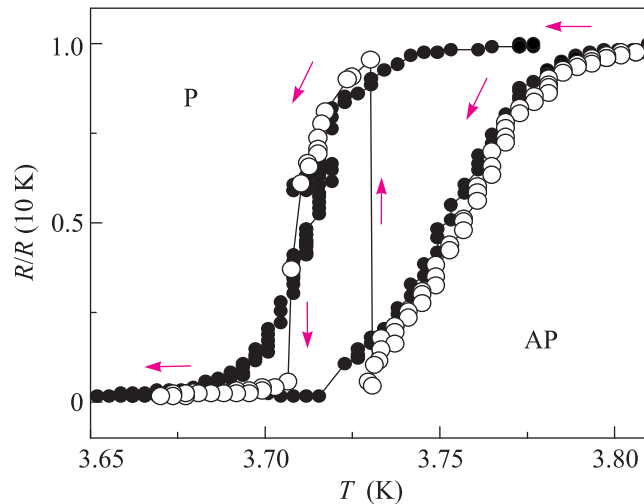

Fig. 4. Superconducting transition curves for P ($H_0 = +1\,\mathrm{kOe}$) and AP ($H_0 = -1\,\mathrm{kOe}$) orientations of the Fe1 and Fe2 layers' magnetizations, respectively, for the sample $CoO_x/Fe1(0.8$ nm$)/Cu(4$ nm$)/Fe2(0.5$ nm$)/Cu(1.2$ nm$)/Pb(60$ nm$)$ (•). Instant switching between superconducting state and normal state by switching between AP ($H_0 = -1\,\mathrm{kOe}$) and P ($H_0 = +1\,\mathrm{kOe}$) orientations of the Fe1 and Fe2 layers' magnetizations during a slow temperature sweep (○)

Let us discuss the results shown in Figs. 2 and 3 in the framework of the S/F proximity effect theory. For that we extend the results of [14] where T_c of an F1/F2/S trilayer was considered in the simplest formulation: all the interfaces were assumed to be transparent, and the outer F layer was assumed to be half-infinite. That formulation enabled studying interplay of different

magnetizations' orientations in the two F layers. For the present experiment, we are interested in theoretical results only for the two collinear orientations (parallel and antiparallel). At the same time, we should extend the previous theory [14] for arbitrary thickness of the outer F layer (Fe1 layer in notation of the present paper).

In this theory, the set of equations describing superconductivity in all parts of the structure is reduced to the problem for the S layer only. All the information about the rest of the structure (two F layers and interfaces) is then contained in a real-valued parameter W that enters the effective boundary condition for the F2/S interface: $\xi(df_0/dx) = Wf_0$, where f_0 is the singlet component of the anomalous Green function in the S layer and ξ is the coherence length. Physically, W determines how strongly superconductivity in the S layer is suppressed by the rest of the structure due to the proximity effect. The larger W is, the stronger T_c is suppressed. Therefore, ΔT_c should correlate with $\Delta W = W^P - W^{AP}$. Note at the same time that generally there is no simple explicit relation (like, e.g., proportionality) between them.

In the parallel configuration, we effectively have F/S system with a single F layer of thickness $d_{Fe1} + d_{Fe2}$. Then we reproduce the result of [20]:

$$W^P = 2k_h\xi\frac{\sigma_F}{\sigma_S} \times$$
$$\times \frac{\cosh(d_1 + d_2) - \cos(d_1 + d_2)}{\sinh(d_1 + d_2) - \sin(d_1 + d_2) + 2\kappa\tanh(k_\omega d_S)}, \quad (1)$$

where

$$d_1 = 2k_h d_{Fe1}, \quad d_2 = 2k_h d_{Fe2}, \quad \kappa = \frac{\sigma_S k_\omega}{\sigma_F k_h}. \quad (2)$$

Here σ_S and σ_F are the normal-state conductivities of the S and F layers, respectively (the diffusion constants will be denoted D_S and D_F); $\xi = \sqrt{D_S/2\pi T_{cS}}$ is the coherence length for the S layer (where T_{cS} is the critical temperature in the bulk). The wave vectors $k_\omega = \sqrt{2\omega/D_S}$ and $k_h = \sqrt{h/D_F}$ describe the scales of the spatial inhomogeneity due to the proximity effect in the S and F layers, respectively (ω is the Matsubara frequency, which can be taken for estimates as πT_c). We assume that T_c/h is small enough so that $\kappa \ll 1$.

In the antiparallel configuration, we generalize treatment of [14] taking into account finite d_{Fe1} (and also taking into account different degree of disorder in the S and F layers, so that D_S, σ_S and D_F, σ_F are different). The result is

$$W^{AP} = 2k_h\xi\frac{\sigma_F}{\sigma_S}\frac{\mathcal{N}}{\mathcal{D}}, \quad (3)$$

where the numerator and denominator of the last fraction are

$$\mathcal{N} = \cosh d_1 \cosh d_2 - \cos d_1 \cos d_2 -$$
$$- \sin d_1 \sinh d_2 - \sinh d_1 \sin d_2, \quad (4)$$
$$\mathcal{D} = \cosh d_1 \sinh d_2 + \sinh d_1 \cos d_2 -$$
$$- \sin d_1 \cosh d_2 - \cos d_1 \sin d_2 + 2\kappa\tanh(k_\omega d_S). \quad (5)$$

The qualitative correlation between measured ΔT_c and calculated ΔW in Figs. 2 and 3 looks satisfactory. Note that $\Delta T_c(d_{Fe2})$ and $\Delta W(d_{Fe2})$ in Fig. 2 approximately correspond to the limit of infinite d_{Fe1} since this thickness is several times larger than the penetration depth $k_h^{-1} \equiv \xi_F$. The central result of our paper, the dependence of ΔT_c on d_{Fe1}, is shown in Fig. 3. For the calculated curves of ΔW we used the same parameters as in our previous paper [18]: for the F layers we put the Fermi velocity $v_F = 2 \cdot 10^8$ cm/s, mean free path of conduction electrons $l_f = 1.5$ nm, and the exchange field $h = 0.85$ eV. According to our calculations $\kappa = 0.009$. The dirty-limit theory (the Usadel equations) that we used to calculate ΔW, requires, in particular, the condition $hl_f/\hbar v_F \ll 1$ (where $\hbar$ is Planck's constant)[3]. The above-mentioned parameters correspond to the value of $hl_f/\hbar v_F$ of the order of one. There are also other simplifications of the theory compared to the experiment; the main of them is probably that all interfaces are assumed to be fully transparent (while the exchange splitting of the conduction band of the ferromagnet is at least one source for a non-perfect F/S interface transparency; see, e.g., [19]). Taking this into account, we can only expect qualitative agreement between theory and experiment.

Let us now discuss our qualitative understanding of the spin-valve effect dependence on d_{Fe1}, keeping in mind that k_h^{-1} is the penetration depth of superconducting correlations in the ferromagnet. In the parallel orientation, the two F layers act as a singe layer leading to some suppression of superconductivity in the S part. The spin-valve effect is due to partial mutual compensation of the exchange fields of the two F layers when they are in the antiparallel orientation. We expect that the spin-valve effect is largest when this compensation is most effective, which is the situation at $d_{Fe1} \sim d_{Fe2}$ if $d_{Fe2} < k_h^{-1}$. At $d_{Fe2} > k_h^{-1}$ this condition should be modified since the outer layer of the same thickness, $d_{Fe1} > k_h^{-1}$ cannot fully participate in the compensation effect. Only a part of it, with thickness of the order of k_h^{-1}, is effective, so we expect the spin-valve effect in this case to be strongest for $d_{Fe1} \sim k_h^{-1}$. The theoretical results for ΔW presented above confirm this

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[3]Note that although the Usadel equations are formally not valid in the case of strong ferromagnets, it turns out that often they can describe experimental results surprisingly well (see, e.g., [8]).

qualitative picture, and the experimental data in Fig. 3, corresponding to $k_h^{-1} \approx 0.8\,\text{nm}$, seem to be consistent with it. Note that the theory also predicts peculiarities of the spin-valve effect at very small d_{Fe1} (due to interference features of the oscillating proximity effect in the F part), however we do not focus on them since we do not have experimental data for such small thicknesses.

In conclusion, we employed a spin valve system $\text{CoO}_x/\text{Fe1}/\text{Cu}/\text{Fe2}/\text{Cu}/\text{Pb}$ to investigate the dependence of the magnitude of the spin valve effect ΔT_c on the thickness of the Fe1 layer d_{Fe1}. We have observed that the ΔT_c value can be slightly (by $\sim 20\%$) increased in comparison with the case of the half-infinite Fe1 layer considered by the theory [14]. The achieved theoretical understanding of the $\Delta T_c(d_{\text{Fe1}})$ dependence warrants a next step towards practical application. Indeed, the optimal choice of the thicknesses d_{Fe1} and of d_{Fe2} gives a possibility to get an almost full switching from the normal to the superconducting state.

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