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Observation of the 'inverse' spin valve effect in a Ni/V/Ni trilayer system

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An experimental study of magnetic and superconducting properties of a trilayer Ni/V/Ni thin film system grown on single-crystalline MgO(001) substrate is reported. The field dependence of the superconducting transition temperature T_c for samples comprising Ni layers with similar values of the coercive field H_c reveals no anomalies. However, in samples with different thicknesses of the nickel layers the difference in H_c amounts up to $\Delta H_c \sim 1.8$ kOe, thus enabling to manipulate the relative orientations of the layers' magnetization by an external magnetic field. Surprisingly, for these samples the T_c for the parallel orientation of the magnetizations of the Ni layers is higher, in a certain magnetic field range, than for the antiparallel one, at odds with theoretical predictions. Possible reasons of this contradiction are discussed.

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The interplay between the two antagonistic ordered states, superconducting and ferromagnetic, in superconducting/ferromagnetic (S/F) multilayer thin film heterostructures has been a subject of intense investigations in the past 15 years (see, e.g., [1]). At present, the interest to this field is further increased in light of theoretical predictions of a possible application of such heterostructures as a spin valve for a superconducting current in spintronic devices [2 – 4]. According to estimates, the superconducting Cooper-pair breaking effect, and hence the superconducting transition temperature T_c, can be controlled by a mutual orientation of the magnetizations of the F-layers under a certain combination of F- and S-layers. In 1999, Tagirov [3] and Buzdin et al. [4] have theoretically suggested a spin valve based on a trilayer F/S/F thin film structure, where the S-layer is sandwiched between two F-layers. Calculations show that an F/S/F trilayer should have a lower T_c for the parallel (P) orientation of the F-layers' magnetization as for the antiparallel (AP) orientation. The reason is that the mean exchange field from two F-layers acting on Copper pairs in the S-layer should be smaller for the AP orientation compared to the P case. The first report on a possible realization of the superconducting spin valve of such design on the basis of a CuNi/Nb/CuNi trilayer system has been published by Gu et al. [5]. The difference in T_c between AP and P orientations of the magnetization in the two CuNi layers ΔT_c turns out to be about 6 mK, whereas the width of the superconducting transition amounts to 50 mK. One should note that the observed effect is not that big, considering the broadness of the superconducting transition and that the T_c of the studied sample was about 3 K. The effect of a similar size for this system has been reproduced later by Potenza et al. [6]. Calculations by [6, 7] show that the experimentally observed shift of T_c is pretty much close to a theoretical limit for this system. This limit is primarily set up by the suppression of superconductivity in the S-layer with a thickness d_s significantly larger than the superconducting coherence length ξ_s, namely when $d_s \sim 3\xi_s$. Later on, there have been several further publications on the observation of the spin valve effect in S/F multilayer thin film systems [8 – 10] that reported record magnitudes of the effect. For instance, Moraru et al. [8] have achieved $\Delta T_c = 41$ mK for the system Ni/Nb/Ni and $\Delta T_c = 20$ mK for the system Py/Nb/Py (Py stands for permalloy). However, a broad superconducting transition width has prevented a full switching between normal and superconducting states. Novak et al. [10] have reached even $\Delta T_c \sim 200$ mK for the sample with a rather thick superconducting V layer grown on an antiferromagnetically coupled [Fe/V] superlattice. However, such a design could not be used so far as a spin valve since it requires quite a strong field to overcome the antiferromagnetic coupling in order to mutually rotate the magnetization of the Fe layers.

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In the present work we have studied superconducting properties of a Ni/V/Ni system with a thickness of the V layer that 4.5 times exceeds ξ_s. To our surprise in some samples we observed the spin valve effect *inverse* to that predicted by theories, namely that the T_c for the parallel orientation of magnetizations is higher than for the antiparallel. Such an 'inverse' superconducting spin valve effect occurs for the samples having a substantial difference in coercive fields of the magnetic layers that enables a manipulation of the mutual orientation of their magnetizations by an external magnetic field.

The Ni/V/Ni samples have been grown by molecular beam epitaxy. First, a single crystalline MgO (001) substrate was heated for 1 hour at a temperature of 600 °C and a pressure of 10^{-6} mBar in a fore-chamber. Next, the substrate was moved into a main chamber where it was kept for 5 minutes at 1000 °C at a base pressure 10^{-11} mBar. The deposition using an electron beam gun took place at 300 °C and 10^{-10} mBar. The growth rate of the vanadium layer amounted to 0.15 nm/s. The nickel layers were grown at a rate of 0.025 nm/s. Finally, a 4 nm palladium protection layer was deposited on the sample. The thicknesses have been determined by a small angle x-ray scattering. It turns out that in some samples the real thickness of Ni layers differs substantially. From sample to sample the thickness of the bottom and top Ni layers varies between 1.6 to 3.2 nm and 3 to 4 nm, respectively. The thickness of the V layer amounts to 44 nm. The roughness of the interface is of the order of 1 nm.

Magnetization of the samples was measured with a SQUID magnetometer. The superconducting transition temperature T_c, the ratio of the resistance values at room temperature and at T_c $RRR = R(300K)/R(T_c)$, as well as the upper critical field H_{c2}, have been determined with a standard four-point contact method in a ^{4}He magnetocryostat. For measurements of H_{c2} magnetic field was applied perpendicular to the direction of the electrical current.

A representative superconducting transition curve measured in a constant magnetic field is shown in Fig.1. One can see that the resistivity starts to decrease at temperatures well above the real superconducting transition. Possibly such a peculiar shape of the superconducting transition is caused by strong fluctuations in the sample core where superconductivity is suppressed by the exchange field.

The magnetic hysteresis loops have been measured at 15 K with the magnetic field direction parallel to the sample plane. A 'usual' hysteresis loop obtained for two studied samples is shown in Fig.2a. A calculation according to [11] reveals similar coercive fields H_c for both Ni layers amounting to 0.19 kOe. An 'anomalous' hysteresis loop which with minor deviations is representative for the other two studied samples is shown in Fig.2b.

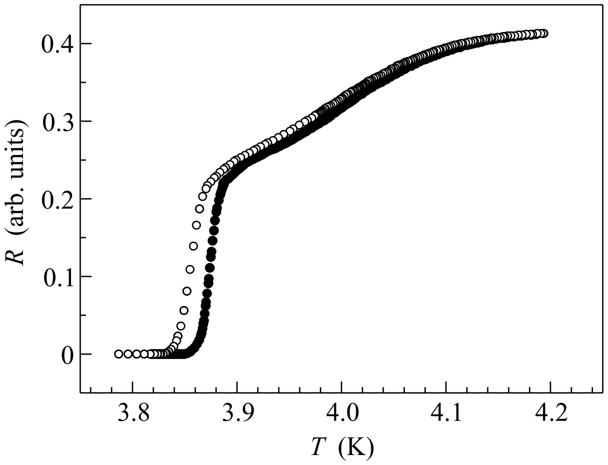

Fig.1. Superconducting transition curves for the sample Ni(1.8 nm)/V(44 nm)/Ni(4 nm) measured in a field of +2 kOe (filled circles) and −2 kOe (open circles). For measurement details see the text

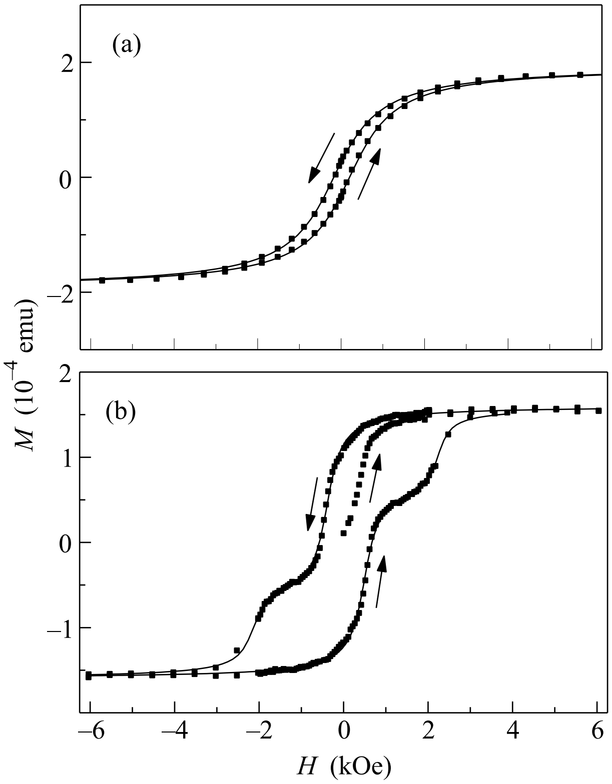

Fig.2. Magnetic hysteresis loops of the samples (a) Ni(3 nm)/V(44 nm)/Ni(3 nm) and (b) Ni(1.8 nm)/V(44 nm)/Ni(4 nm). Magnetic field is directed in the sample plane. Solid lines are theoretical fits

With an initial increase of the magnetic field from zero the magnetization of both Ni layers increases and saturates at a field of about 6 kOe. A subsequent decrease of the field to zero and sweeping it between negative and positive values yields characteristic steps in the hysteresis loop indicative of different coercive fields of the two Ni layers. A fit of the data using the algorithm from [11] shows that the coercive fields H_c of the 'soft' and 'hard' Ni layers amount to 0.44 and 2.2 kOe, respectively, their magnetization ratio being of the order of 2.3. All H_c values obtained for the studied samples are considerably larger than those expected for a 'free' Ni layer. The obtained difference in the coercive fields of the two F-layers is, apart from different growth conditions on the MgO substrate and on the V layer, related to their different thicknesses. It is well known that the coercive field increases with decreasing the F-layer thickness. Recalling that the thickness of the bottom Ni layer is found to be systematically smaller than that of the top Ni layer, we can assign a coercive field of 2.2 kOe to the bottom F-layer deposited directly on the MgO substrate, and a coercive field of 0.44 kOe to the top F-layer deposited on the V surface. This condition enables to manipulate a mutial orientation of the magnetizations of the F-layers by changing an external magnetic field in a range ±1 kOe.

The dependence of the upper critical field H_{c2} on temperature for parallel and perpendicular orientations of a magnetic field relative to the sample plane is shown in Fig.3. They are similar for all samples. The $H_{c2}(T)$ curves can be qualitatively described by known expressions for superconducting thin films [12]. For the perpendicular orientation H_{c2} is linear in T, whereas in the parallel case it follows a $\sqrt{(1-T/T_c)}$ dependence. For samples with a 'normal' hysteresis loop the $H_{c2}(T)$ dependence has a parabolic behavior shown in Fig.3 for an arbitrary field direction, whereas for samples with an 'anomalous' hysteresis loop a close look reveals a difference in experimental data depending on the direction of a magnetic field in the sample plane (see Fig.4).

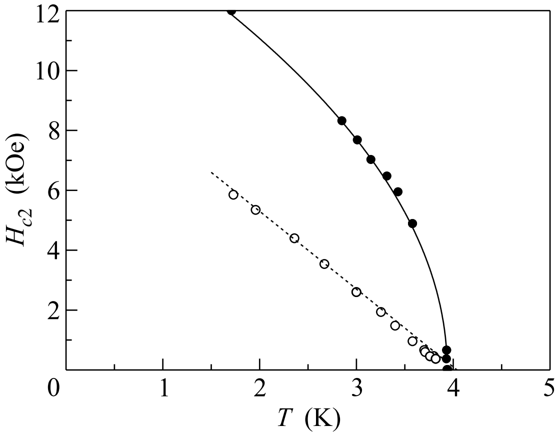

Fig.3. Temperature dependence of the upper critical field H_{c2} of the samples for the parallel (filled circles) and perpendicular (open circles) orientation of the external magnetic field with respect to the sample plane. Solid line represents the dependence $H_{c2} = 7.95\sqrt{(1-T/3.94)}$. Dashed line is a linear approximation of H_{c2} for the perpendicular orientation of the field

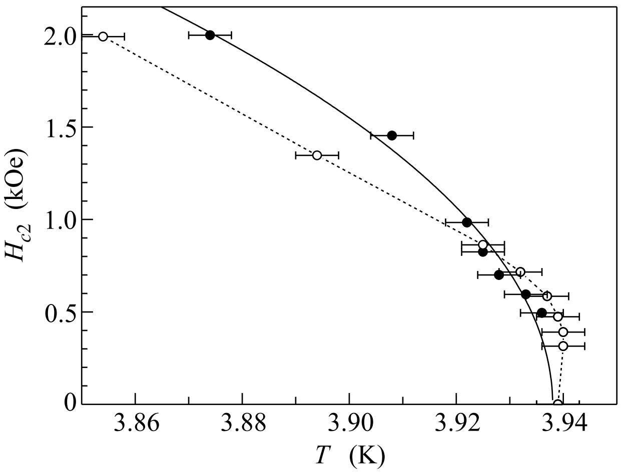

Fig.4. $H_{c2}(T)$ dependence of the sample Ni(1.8 nm)/V(44 nm)/Ni(4 nm) for the orientation of the magnetic field parallel to the sample plane. Filled and open circles correspond to the positive and negative in-plane field directions, respectively. Solid line is the same parabolic dependence as shown in Fig.3, dashed line is a guide for the eye. At $T \simeq 3.8$ K both curves merge. For measurement details see the text

The value of $RRR = R(300K)/R(T_c)$ for the studied samples is found to be of the order of 5. From this value, using the approach described before in [13], we have estimated the superconducting coherence length ξ_s ~10 nm which is approximately a quarter of the S-layer thickness. From calculations in Ref. [7] we may conclude that $\Delta T_c = T_c^{AP} - T_c^{P}$ rapidly decreases with increasing d_s. For our actual thickness of ~44 nm it can be estimated as 3–5 mK, assuming the equal F-layer thickness.

The study of superconducting properties of the Ni/V/Ni samples with an 'anomalous' magnetic hysteresis loop has led to an observation of a phenomenon inverse to the spin valve effect. Specifically, in the case of the AP orientation of the magnetizations the T_c is found to be smaller compared to the P case.

The spin valve effect has been studied by measuring the $H_{c2}(T)$ dependencies close to T_c for different orientations of the field direction in the sample plane in the following way: Initially, a magnetic field applied in the film plane was set to 6 kOe which corresponds to a magnetic saturation of the samples and thus to the P case. Then the field strength was lowered down to 2 kOe. After that the field was further gradually decreased and T_c was measured at fixed field values (see Fig.4). After crossing the zero and increasing the field strength in the opposite direction the magnetizations of the F-layers begin to misalign (see Fig.2b). Up to the field of -0.7 kOe ΔT_c between this misaligned and P orientation is positive and amounts not more than 5 ± 4 mK. In spite of a good agreement with the estimate given above, it is obviously within the error bar, considering the width of the superconducting transition of 40 mK. Further increase of the field in the range -1 to -2 kOe yields the superconducting transition temperature smaller than T_c^P. The difference in T_c in the negative field range 1.5 - 2 kOe (here the magnetization direction is close to the AP case) achieves the value $T_c - T_c^P = -20 \pm 4$ mK. Finally, at a field of -3 kOe the T_c curves merge. Note, that the magnitude of ΔT_c is significantly larger compared to theoretical predictions for our layer thicknesses and, moreover, it has the opposite sign. Thus, as it is seen from Fig.4, at the magnetic field range between -0.7 and $+0.7$ kOe we observe a 'normal' spin valve effect; out of this range up to a magnetic field of ± 3 kOe an 'inverse' spin valve effect is found.

Earlier, a similar effect has been observed in trilayer systems YBaCuO/LaSrMnO/YBaCuO [14] and Py/Nb/Py, (Py=$Ni_{80}Fe_{20}$) [15]. Rusanov et al. [15] have pointed out that the superconducting layer in their system is in a close contact with a strong ferromagnet which could cause an 'inverse' spin valve effect. They have suggested the following explanation: Assume a superconducting current flowing in the sample plane. Electrons entering the S-layer from the first F-layer form spin polarized quasiparticles with the energy of the order or smaller than the superconducting gap, since there is no potential difference in the perpendicular direction. Some of them scatter back into the F-layer due to the wave vector mismatch, others diffuse through the S-layer. In contrast to the P case, in the AP configuration most of the latter quasiparticles will be reflected back from the second F-layer because their spin polarization is opposite to the magnetization of that layer. Accumulation of the spin polarized quasiparticles in the superconducting volume should result in the suppression of the gap and hence of superconductivity. The question arises, why the 'inverse' spin valve effect takes place only in the F/S/F samples with a significantly thick S-layer? Note, that Moraru et al. [8], who have studied the Ni/Nb/Ni and Py/Nb/Py trilayers with a much thinner Nb layer system, have observed, as mentioned above, the 'normal' spin valve effect with one of the record values of ΔT_c. There, the effect is maximum for thicknesses of the S-layer close to a critical value below which superconductivity disappears.

It is unlikely that the anomalous behavior of T_c in our samples could be due to the dipolar fields from domain boundaries [16]. Most probably, in our samples the Néel type domains are realized that should not give rise to dipolar fields in the S-layer. Furthermore, the influence of the dipolar fields in the Ni/Nb/Ni system with the thickness of the Nb layer of 17–18 nm [8] should be significantly stronger than in our Ni/V(44 nm)/Ni system, which is not observed experimentally.

Finally, along with the singlet superconductivity, one cannot exclude the occurrence of the long-range triplet pairing in F/S/F structures. It may arise due to a noncollinear configuration of magnetizations in the F-layers [17], that could be possibly realized in our samples close to the switching to the AP configuration. Explicit calculations for F/S/F trilayers accounting for the triplet pairing have shown that T_c^{AP} for the antiparallel configuration is always the highest superconducting transition temperature [18]. This conclusion is in contrast with our observation of the 'inverse' spin-valve effect in Ni/V/Ni trilayers.

In summary, by studying magnetic and superconducting properties of the Ni/V/Ni system we have found a surprising 'inverse' proximity effect in a certain temperature and magnetic field range, that yields a higher superconducting transition temperature in the V layer for the parallel orientation of the magnetizations in the ferromagnetic Ni layers. Our experimental observation should motivate further studies to elucidate the reasons of this effect and thus makes up an important step towards realization of the spin valve for the superconducting current.

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