

Superconducting Spin Valve Effect of a V Layer Coupled to an Antiferromagnetic [Fe/V] Superlattice

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We studied superconducting V layers deposited on an antiferromagnetically coupled $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice. The parallel upper critical magnetic field exhibits an anomalous T dependence up to the ferromagnetic saturation field of the superlattice, indicating that the superconducting transition temperature T_S decreases when rotating the relative sublattice magnetization directions of the superlattice from antiparallel to parallel. This proves that the pair breaking effect of a Fe_2 layer is reduced if at a distance of 1.5 nm a second Fe_2 layer with antiparallel spin orientation exists.

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The interplay between magnetism and superconductivity in thin film systems is a fascinating phenomenon which attracts increasing interest in recent literature [1,2], since exotic superconducting states can exist here and novel superconducting phenomena can be expected. The existence of π -wave superconductivity in ferromagnetic/superconductor (F/S)-layer systems is now well established [3,4]; the propagating character of the superconducting pair wave function in F/S systems leading to characteristic oscillations in the superconducting transition temperature as a function of the thickness of the ferromagnetic layer also has been detected experimentally [5,6]. There are other interesting theoretical predictions for S/F systems which wait for experimental realization, e.g., the occurrence of triplet pairing [7,8] or the inverse proximity effect [9].

An interesting situation occurs in $F_1/S/F_2$ systems where the superconducting layer is in contact with two different ferromagnetic layers F_1 and F_2 whose magnetization directions can be reversed independently. Recent model calculations showed that the superconducting transition temperature T_S should be lowered if the relative magnetization direction of F_1 and F_2 is rotated from antiparallel to parallel [10,11]. Since this effect can be used to switch the superconductivity on and off by reversing the magnetization direction of a ferromagnetic film, this has been dubbed the superconducting spin valve effect [10]. The basic reason for this effect is that the strong pair breaking of Cooper pairs by the ferromagnetic exchange field is partly cancelled if the magnetization direction of F_1 and F_2 is antiparallel.

A prerequisite for the observation of a sizable superconducting spin valve effect for $F/S/F$ trilayers is that the superconducting correlation length ξ_S is of the order or

larger than the separation of the two ferromagnetic layers [10,11]. Experimentally it turned out that this condition is very difficult to meet in combinations of elementary superconductors like Nb, V, or Pb with ferromagnetic transition metals. In the standard trilayer system, with a ferromagnetic film on the bottom and the top of a superconducting film, the pair breaking effect of even a very thin ferromagnetic film is so strong that the critical thickness for the onset of superconductivity shifts to values $d_S > 4\xi_S$ [5,6]. There is only one recent report in the literature on the successful realization of a superconducting spin valve using a CuNi/Nb/CuNi trilayers system [12]. However, the maximum shift of the superconducting transition temperatures ΔT_S for the magnetization of the CuNi layers being either parallel or antiparallel was only 6 mK, or about 0.2% of T_S . Actually, this shift is of the same order of magnitude where the domain structure of the ferromagnetic layers has an influence on T_S .

We have taken a different approach for the realization of the superconducting spin valve effect which is based on the theoretical work in Ref. [13] and has not been realized experimentally until now. This is the $S/F_1/N/F_2$ layer scheme where two ferromagnetic layers F_1 and F_2 separated by a thin nonmagnetic (N) layer are deposited on one side of the superconductor with F_1 and N thin enough to allow the superconducting pair wave function to penetrate into F_2 . Then, rotating the relative magnetization direction of F_1 and F_2 from parallel to antiparallel the model calculations gave a substantial difference ΔT_S assuming reasonable microscopic parameters for the superconducting and ferromagnetic films.

In our experimental realization of the $S/F_1/N/F_2$ layer scheme, the ferromagnetic layers F_1 and F_2 are 2 monolayers of Fe (Fe_2) separated by 11 monolayers of V (V_{11})

and grown epitaxially on a thick superconducting V film (see Table I). The V_{11} layer mediates an antiferromagnetic interlayer exchange coupling (IEC) between the Fe_2 layers [14,15] so that one can rotate the relative magnetization direction of F_1 and F_2 from antiparallel to parallel in an external field and observe the shift in the transition temperature ΔT_S . For practical purposes, we used an antiferromagnetically coupled epitaxial superlattice $[Fe_2V_{11}]_{20}$ [15] instead of one single $Fe_2/V_{11}/Fe_2$ trilayer. For the superconducting proximity effect this makes essentially no difference, since only the two ferromagnetic layers closest to the thick superconducting V layer are relevant.

There are several reasons which make the epitaxial (V/Fe) system a favorable choice for demonstrating the superconducting spin valve effect: First, it is the superior quality of the Fe/V interface in the superlattice [16] which guarantees a high interface transparency and weak diffusive pair breaking scattering at the interface. Second, the Fe_2 layers have a thickness of only about 0.3 nm, and the exchange splitting I_0 of the conduction band is reduced by about a factor of 2 compared to bulk Fe, as the number of Fe nearest neighbors is reduced by this factor. The decay length of the superconducting pair density in a ferromagnetic layer given by $\xi_F = (4\hbar D_F/I_0)^{1/2}$ (D_F being the conduction electron diffusion constant in the ferromagnet) is about 0.8 nm in bulk Fe [6] and will increase to about 1.2 nm in the Fe_2 layers. Thus the pair wave function within the Fe_2 layer will only be weakly damped and the condition $d_F/\xi_F < 0.4$ optimum for observing the superconducting spin valve effect is fulfilled [13].

We have prepared a series of six samples $MgO(100)/[Fe_2V_{11}]_{20}/V(d_V)$, the thickness d_V of the single superconducting V layer was varied between 16 and 30 nm (Table I). An Al_2O_3 -cap layer with a thickness of 2 nm was deposited as a protection against oxidation. The $[Fe_2V_{11}]_{20}(100)$ superlattice was grown epitaxially on $MgO(100)$, as described in detail elsewhere [17]. The properties of these (Fe/V) superlattices have been studied

TABLE I. Parameters of the samples of the present study with the design $MgO(100)/[Fe_2V_{11}]_{20}/V(d_V)$ and the thickness of the superconducting V layer d_V , the residual resistivity ratio defined as $RRR = R(300\text{ K})/R(10\text{ K})$, the superconducting correlation length ξ_S , the ratio d_V/ξ_S , the superconducting transition temperature T_S , and the shift of the superconducting transition temperature between the Fe_2/V_{11} superlattice in the antiferromagnetic state and in ferromagnetic saturation ΔT_S .

d_V (nm)	RRR	ξ_S (nm)	d_V/ξ_S	T_S (K)	ΔT_S (K)
16	3.0	5.9	2.71	1.79	-0.10 ± 0.01
18	4.1	7.2	2.50	2.07	-0.12 ± 0.01
20	4.2	7.3	2.73	2.67	-0.06 ± 0.02
22	3.5	6.6	3.33	2.58	-0.04 ± 0.01
26	6.5	9.7	2.68	3.33	-0.07 ± 0.02
30	7.8	10.8	2.77	3.62	-0.11 ± 0.01

thoroughly during the past few years [15–18], the Fe layers are ferromagnetic down to a thickness of 2 Fe monolayers, and there is only a very narrow V-thickness range with antiferromagnetic interlayer exchange coupling which we need here [15].

The superconducting transition temperature and the upper critical magnetic field $H_{c2}(T)$ for the direction parallel and perpendicular to the film plane were measured resistively by standard four point dc technique. The magnetic field is generated by superconducting Helmholtz coils and the film can be rotated *in situ* relative to the magnetic field direction with a high precision, which is important for the direction of the field axis parallel to the film plane. The magnetic hysteresis loops are measured by a commercial SQUID magnetometer (Quantum Design MPMS-system).

In Fig. 1 we reproduce the magnetization curve of the $[Fe_2V_{11}]_{20}$ superlattice measured at 10 K. The shape of the hysteresis shows that the IEC is antiferromagnetic with a ferromagnetic saturation field of $H_{sat} = 6$ kOe. The ferromagnetic saturation magnetization corresponds to a magnetic moment of $0.38 \mu_B/Fe$ atom. The reduced Fe moment is mainly due to induced V-magnetic moments at the interface with a moment direction antiparallel to the Fe moments [19]. The magnetization curve in Fig. 1 exhibits some hysteresis with a small ferromagnetic remanence, indicative of a not completely perfect antiferromagnetic alignment of the Fe_2 layers. The magnetic hysteresis loops are virtually identical for all superlattices investigated.

The resistive transition with the magnetic field applied parallel to the film plane has a width of 0.1 K without any observable broadening in high magnetic fields, as expected theoretically for a thin film if no vortices enter. We define the upper critical field $H_{c2}(T)$ by taking the 50% value of the resistive transition. The upper critical field for the magnetic field direction parallel and perpendicular to the

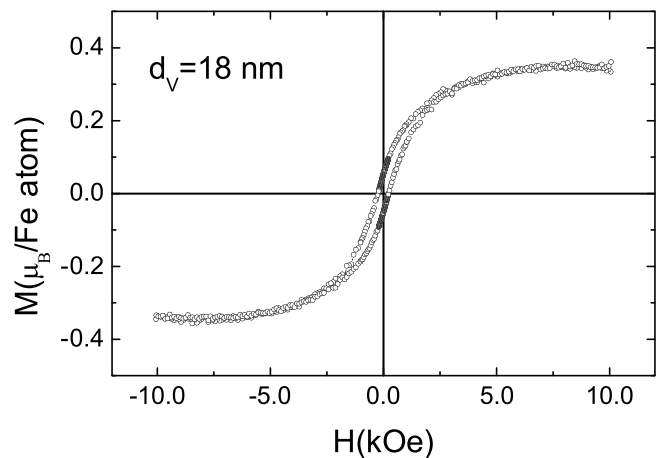


FIG. 1. Magnetic hysteresis loop of the sample $[Fe_2V_{11}]_{20}/V(18\text{ nm})$ measured at 10 K.

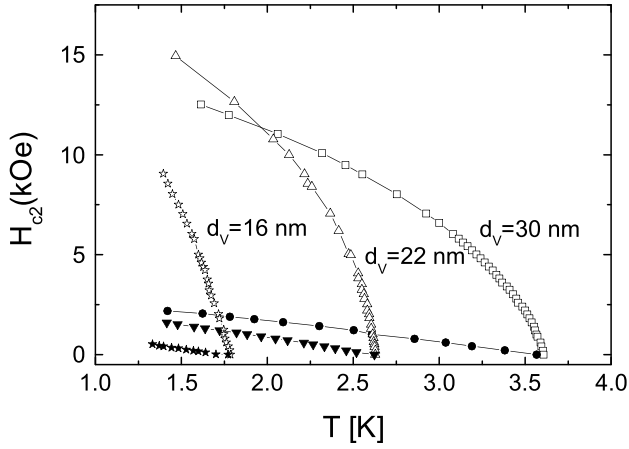


FIG. 2. Upper critical magnetic field versus temperature with the field applied parallel and perpendicular to the film plane for three samples $[\text{Fe}_2\text{V}_{11}]_{20}/\text{V}(d_V)$ from Table I. The thickness d_V is given in the figure, the open symbols refer to the magnetic field direction parallel to the film plane, the solid symbols refer to the direction perpendicular to the plane.

film plane is plotted in Fig. 2 for several samples from Table I. For a two dimensional (2D) thin film with the magnetic field parallel or perpendicular to the film plane the classical result for the upper critical field is [20]

$$H_{c2}^{\text{perp}} = \frac{\Phi_0}{2\pi\xi^2(0)} \left(1 - \frac{T}{T_S}\right), \quad (1a)$$

$$H_{c2}^{\text{par}} = \frac{\Phi_0}{2\pi\xi(0)} \frac{\sqrt{12}}{d_S} \sqrt{\left(1 - \frac{T}{T_S}\right)}, \quad (1b)$$

with the flux quantum Φ_0 , the thickness of the film d_S , and the Ginzburg-Landau correlation length ξ , which is related to Pippard's correlation length listed in Table I by $\xi(0) = 1.6\xi_S$. The typical 2D temperature dependence formula (1b) is strictly valid if the condition $d_S < 1.84\xi(T)$ holds [21]. With a ferromagnetic film at the surface of the superconducting film, the limit for d_S shifts to definitely higher values [22]; thus all of our films in Table I are in the 2D limit. The prefactors in formulas (1a) and (1b) do not hold exactly for a 2D film in contact with a ferromagnetic film; there are corrections which can only be obtained numerically [22]. We have performed measurements of the upper critical field for Fe/V/Fe trilayers and obtained an $H_{c2}^{\text{par}}(T)$ in perfect agreement with formula (1b), similar to what has been observed earlier [23,24].

In Figs. 3(a) and 3(b) we have plotted the square of the parallel upper critical field on an enlarged scale together with the straight line which describes the temperature dependence for fields above 6 kOe perfectly. Below $H = 6$ kOe there is an increasing deviation from the straight line. From the extrapolation of the straight line, one gets a superconducting transition temperature T_S which is more than 0.1 K below the true transition temperature measured

at zero field. The shape of the deviation is virtually identical for all samples from Table I, only the amplitude is different. A comparison with the magnetization curve of the $[\text{Fe}_2/\text{V}_{11}]$ superlattice in Fig. 1 shows that the ferromagnetic saturation field of 6 kOe is correlated with the first deviation of H_{c2}^{par} from the straight line in Figs. 3(a) and 3(b). From this we infer that the deviation of the upper critical field from the 2D behavior in Fig. 3 is caused by the gradual change of the sublattice magnetization direction of the $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice from parallel above 6 kOe to antiparallel in zero field. For the sample with $d_V = 16$ nm in Fig. 3(a) the superconducting transition temperature in the antiferromagnetic state is $T_S = 1.78$ K, while in ferromagnetic saturation we extrapolate $T_S = 1.67$ K. The temperature difference $\Delta T_S = 0.11$ K is the anticipated

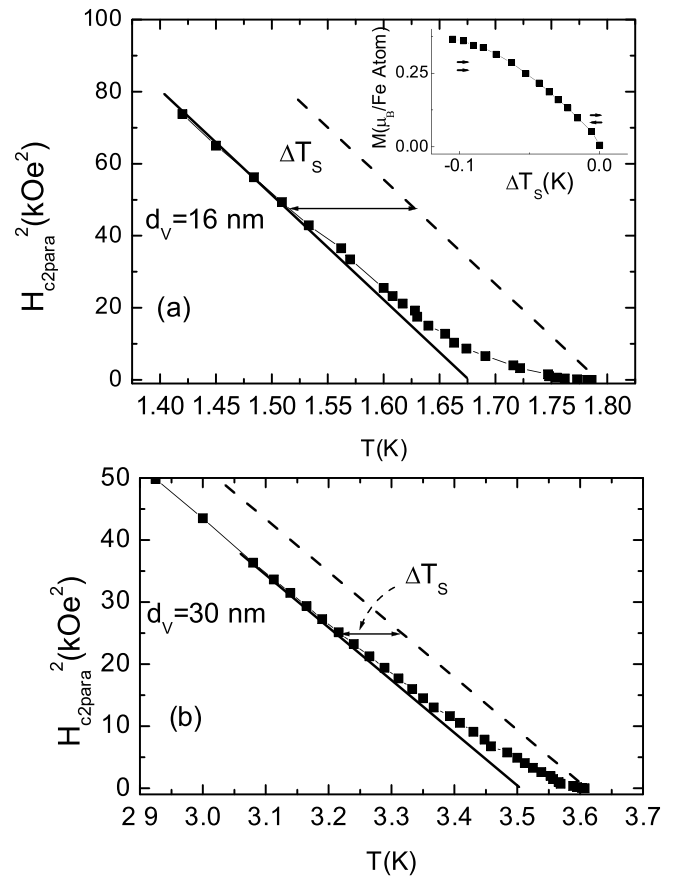


FIG. 3. Square of the parallel upper critical magnetic field versus temperature for the sample $[\text{Fe}_2\text{V}_{11}]_{20}/\text{V}(16 \text{ nm})$ (a) and $[\text{Fe}_2\text{V}_{11}]_{20}/\text{V}(30 \text{ nm})$ (b). The solid straight line is the extrapolation of the linear temperature dependence for higher fields, and the dashed line is the theoretical curve expected if the magnetization of the superlattice does not change. ΔT_S is the shift of the superconducting transition temperature between the superlattice in the antiferromagnetic state and in ferromagnetic saturation. The inset in panel (a) depicts the shift of the superconducting transition temperature with the magnetization of the $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice.

superconducting spin valve effect. This conclusion is corroborated in the inset of Fig. 3(a), where we have plotted the deviation ΔT_S from the linear curve versus the magnetization of the $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice. ΔT_S is largest in the parallel state and decreases continuously towards the antiferromagnetic state, consistent with theoretical predictions [11].

In Table I we summarize the values for ΔT_S of all six samples of the present study together with other parameters which can be derived from our set of experimental data. The superconducting correlation length ξ_s given in the third column of Table I has been calculated using Pippard's dirty limit formula [25] $\xi_s = \sqrt{\xi_0 l / 3.4}$ with the BCS coherence length $\xi_0 = 44$ nm for V and the mean free path l of the conduction electrons derived from the residual resistivity ratio (RRR) which is also given in Table I. The superconducting spin valve effect exists for all samples in Table I; however, it does not simply scale with the thickness of the V film. The reason is that, with all other parameters fixed, ΔT_S should scale with ξ_s/d_S [10,11,13]. The data in Table I roughly reproduce this correlation; however, there are remarkable deviations well above the experimental resolution limit. This indicates that subtle structural modifications of the nominally identical $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattices lead to noticeable changes in ΔT_S . Besides the ξ_s/d_S ratio, the most important factors determining ΔT_S are the interface transparency and the strength of the spin orbit scattering at the F/S interfaces [10]. Since in our geometry the second Fe_2 layer is separated by three interfaces from the thick V layer, slight differences in the interface quality can sum up to cause sizable changes in ΔT_S .

Concluding, our experiments show that the superconducting transition temperature of a V film sensitively reacts on the relative magnetization orientation of the Fe_2 layers of an antiferromagnetically coupled $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice. This system can be turned into a real superconducting switching device by replacing the antiferromagnetically coupled $[\text{Fe}_2\text{V}_{11}]_{20}$ superlattice by a conventional spin valve trilayer system on top of the superconductor. Such a switch would be highly interesting for superconducting logics and data storage.

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- [1] C. L. Chien and D. H. Reich, *J. Magn. Magn. Mater.* **200**, 83 (1999).
- [2] I. A. Garifullin, *J. Magn. Magn. Mater.* **240**, 571 (2002).
- [3] T. Kontos, M. Aprili, J. Lesueur, and X. Grison, *Phys. Rev. Lett.* **86**, 304 (2001).
- [4] V. V. Ryazanov, V. A. Oboznov, A. Yu. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, *Phys. Rev. Lett.* **86**, 2427 (2001).
- [5] L. Lazar, K. Westerholt, H. Zabel, L. R. Tagirov, Yu. V. Goryunov, N. N. Garif'yanov, and I. A. Garifullin, *Phys. Rev. B* **61**, 3711 (2000).
- [6] I. A. Garifullin, D. A. Tikhonov, N. N. Garif'yanov, L. Lazar, Yu. V. Goryunov, S. Ya. Khlebnikov, L. R. Tagirov, K. Westerholt, and H. Zabel, *Phys. Rev. B* **66**, 020505(R) (2002).
- [7] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, *Phys. Rev. Lett.* **86**, 4096 (2001).
- [8] A. F. Volkov, F. S. Bergeret, and K. B. Efetov, *Phys. Rev. Lett.* **90**, 117006 (2003).
- [9] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, *Phys. Rev. B* **69**, 174504 (2004).
- [10] L. R. Tagirov, *Phys. Rev. Lett.* **83**, 2058 (1999).
- [11] I. Baladie, A. Buzdin, N. Ryzhanova, and A. Vedyayev, *Phys. Rev. B* **63**, 054518 (2001).
- [12] J. Y. Gu, C.-Y. You, J. S. Jiang, J. Pearson, Ya. B. Bazaliy, and S. D. Bader, *Phys. Rev. Lett.* **89**, 267001 (2002).
- [13] Oh. Sangjun, D. Youm, and M. R. Beasley, *Appl. Phys. Lett.* **71**, 2376 (1997).
- [14] P. Bruno and C. Chappert, *Phys. Rev. B* **46**, 261 (1992).
- [15] B. Hjörvarsson, J. A. Dura, P. Isberg, T. Watanabe, T. J. Udovic, G. Andersson, and C. F. Majkrzak, *Phys. Rev. Lett.* **79**, 901 (1997).
- [16] G. Andersson, B. Hjörvarsson, and H. Zabel, *Phys. Rev. B* **55**, 15 905 (1997).
- [17] P. Isberg, B. Hjörvarsson, R. Wräppling, E. B. Svedvberg, and L. Hultman, *Vacuum* **48**, 483 (1997).
- [18] V. Uzdin, K. Westerholt, H. Zabel, and B. Hjörvarsson, *Phys. Rev. B* **68**, 214407 (2003).
- [19] V. Uzdin, D. Laberge, K. Westerholt, H. Zabel, and B. Hjörvarsson, *J. Magn. Magn. Mater.* **240**, 481 (2002).
- [20] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1978).
- [21] V. G. Kogan, *Phys. Rev. B* **34**, 3499 (1986).
- [22] Z. Radovic, L. Dobrosavljevic-Grujic, A. I. Buzdin, and J. R. Clem, *Phys. Rev. B* **38**, 2388 (1988).
- [23] H. J. Wong, B. Y. Jin, H. Q. Yang, J. B. Ketterson, and J. E. Hilliard, *J. Low Temp. Phys.* **63**, 307 (1986).
- [24] P. Koorevaar, Y. Suzuki, R. Coehoorn, and J. Aarts, *Phys. Rev. B* **49**, 441 (1994).
- [25] A. B. Pippard, *Rep. Prog. Phys.* **23**, 176 (1960).