Superconducting/ferromagnetic proximity effect mediated by Cr spacer layers

I. A. Garifullin,* D. A. Tikhonov, N. N. Garif'yanov, and M. Z. Fattakhov
Zavoisky Physical-Technical Institute, 420029 Kazan, Russia

L. R. Tagirov
Kazan State University, 420008 Kazan, Russia

K. Theis-Bröhl, K. Westerholt, and H. Zabel
Institut für Experimentalphysik/Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany

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We have studied the superconducting proximity effect in the thin film system Fe/Cr/V/Cr/Fe where the Cr layers play the role of screening layers between the superconducting V-layer and the strongly pair breaking Fe-layers. When keeping the thickness of the Fe-layers d_{Fe} fixed and varying the thickness of the Cr-layers d_{Cr}, the superconducting transition temperature T_c first rises reaching a maximum at $d_{Cr}=40$ Å and then sharply drops for larger Cr-thickness. Keeping d_{Cr} constant and varying d_{Fe} the superconducting transition temperature becomes independent on d_{Fe} for $d_{Cr}>40$ Å. The results demonstrate that the Cooper pairs penetrate into the Cr-layer to a depth of about 40 Å. From our experimental results we suggest that the Cr-layer is nonmagnetic for $d_{Cr}<40$ Å and undergoes a transition to an incommensurate spin density wave state for $d_{Cr}>40$ Å.

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I. INTRODUCTION

The influence of ferromagnetic (F) layers on the superconductivity of superconducting (S) layers in S/F thin film heterostructures is by now well understood through detailed experimental studies of the combinations Nb/Gd,[1,2] Nb/Fe,[3,4] Pb/Fe,[5] $Nb/Cu_{0.43}Ni_{0.57}$,[6] and V/Fe.[7,8] Less extensively investigated are heterostructures consisting of antiferromagnetic (AF) and superconducting layers in S/AF thin film systems. Usually it is assumed that the S/AF proximity effect is identical to the conventional proximity effect at the boundary between normal (N) and superconducting metal layers. This assumption is rationalized by the fact that on the length scale of the superconducting coherence length ξ_s, the exchange field in an antiferromagnet averages out. At low temperatures the Cooper pairs should be able to penetrate deeply into the AF-layer, since the penetration depth in S/N system is given by $\xi_{NM}=\sqrt{\hbar D/2\pi k_B T}$.[9] In contrast, in S/Cr heterostructures a strong suppression of the superconducting transition temperature has been observed, such as in Pb/Cr,[10] Nb/Cr,[11] and V/Cr.[12–18] This has usually been attributed to strong inelastic pair breaking scattering in the Cr-layer by magnetic defects. Furthermore, our recent study of the proximity effect in V/Cr heterostructures[18] revealed a rather unusual thickness dependence of the critical temperature on the Cr layer thickness $T_c(d_{Cr})$, with a strong anomalous suppression of T_c for $d_{Cr}>50$ Å, which we explained by the onset of incommensurate spin density wave order. From these facts it is quite clear that S/Cr heterostructures cannot be treated like normal S/N systems.

It is well known that Cr is an itinerant antiferromagnet displaying an incommensurate spin density wave (ISDW) below the Néel temperature at $T_N=311$ K.[19] The ISDW is a sinusoidal modulation of the antiferromagnetically arranged magnetic moments, with a period increasing from about 60 Å at low temperatures to about 70 Å at the Néel temperature.

In thin films the ISDW magnetism of Cr becomes strongly affected by the respective magnetic or electronic boundary conditions.[20–22] For instance, Fe layers adjacent to Cr confines the ISDW in the direction normal to the ferromagnetic layers and pins the antinodes at the Fe/Cr interface.[21,23] For commensurate antiferromagnetic CrMn boundary layers, this confinement becomes perfect.[24] In contrast, for a vanadium boundary layer a node is expected at the interface as the SDW amplitude is strongly suppressed in V.[23,25] Mibu *et al.*[26,27] have reported Mössbauer spectroscopy experiments showing that the Cr magnetic moment near the V/Cr interface vanishes up to a thickness of 20 Å away from the interface, while at larger distances of above 40 Å the Cr magnetic moment is reestablished, reaching values comparable to the bulk moment. Whether this is a finite size scaling effect as has been observed for Cr confined between Fe layers,[23,28] or whether this is due to hybridization of *d*-bands of V and Cr is not clear at present. In any case, the spin density wave can only develop for Cr films thicker than about 40 Å.

The assumption that the AF-order is irrelevant for the superconducting proximity effect is certainly an oversimplification for Cr. Only for antiferromagnets consisting of localized moments this would be true. But in Cr the SDW-state modifies the spin-up and spin-down states at the Fermi surface and involves the same electrons undergoing the BCS condensation in the superconducting state. In this sense the AF-order and the penetration of the Cooper pairs from the V-layer into Cr can be considered as a competition of two different types of electronic order (see, e.g., Ref. 29).

In the present paper we approach the problem of the V/Cr proximity effect from a different point of view. In V/Fe heterostructures we interleave thin Cr-layers between the V- and Fe-layers with the aim to screen the strong exchange field of the Fe-layer from the Cooper pairs. Simultaneously, however, the different states of Cr, including the onset of magnetic order with increasing thickness, should have an

TABLE I. Layer structure and thickness of the layers in the sample series (1)–(4) used in the present study. Each column presents one sample series.

Series	1	2	3	4
d_{Cr} (Å)-seed layer	40	40	40	40
d_{Fe}(Å)	50	8–20	9–24	8–22
d_{Cr}(Å)	23–52	15	28	47
d_V(Å)	300	300	300	300
d_{Cr}(Å)	23–53	15	28	47
d_{Fe}(Å)	50	8–20	9–24	8–22
d_{Cr}(Å)-cap layer	40	40	40	40

effect on the superconductivity. Both, the screening effect and the intrinsic magnetic ordering effect competing with superconductivity can be separated by a systematic variation of both thicknesses, d_{Cr} and d_{Fe}.

II. SAMPLE PREPARATION AND CHARACTERIZATION

The Fe/Cr/V/Cr/Fe thin films of the present study were grown on single crystalline MgO(001) substrates by molecular beam epitaxy (MBE) with the system having a base pressure below 10^{-9} mbar. Fe and Cr were evaporated from effusion cells with an evaporation rate of about 0.1 Å/s. For the V-layer we used electron beam evaporation and a growth rate of 1.4 Å/s. A growth temperature of 300 °C was chosen for all layers, this temperature representing a good compromise between crystallinity and low interdiffusion at the interfaces. *In situ* reflection high energy electron diffraction (RHEED) analysis during the growth process revealed smooth layer growth of all layers.

In total we prepared four series of samples. The layer thicknesses characterizing each series are listed in Table I. In series (1) the Cr-layer thickness d_{Cr} was varied while keeping the thickness of the Fe-layer $d_{Fe}=50$ Å fixed. In the other 3 series [series (2), (3), and (4)] d_{Cr} was fixed and d_{Fe} changed. The thickness of the V-layer was fixed to 300 Å for all samples. Our previous work on V/Fe (Ref. 8) and V/Cr (Ref. 18) trilayer systems showed that at $d_V=300$ Å one observes a high sensitivity of the superconducting transition temperature T_c as a function of the Cr or Fe-layer thickness.

The four series of samples (Table I) were prepared within one single run. When growing the layers with constant thickness within one series, the substrate holder was rotated. During the growth of the variable layer thickness the substrate holder was positioned asymmetrically with respect to the effusion cell, thus using the natural gradient in the evaporation rate. For all samples a Cr seed layer of 40 Å thickness was the first layer grown on the MgO substrate. In a final step, the samples were covered by a 50 Å thick protective Cr cap layer.

The interface quality and crystal structure were investigated by small angle reflectivity measurements and high angle Bragg scans. Figure 1 reproduces three reflectivity scans for samples from different series, using Mo K_α-radiation ($\lambda=0.709$ nm) and a Si (111) monochromator.

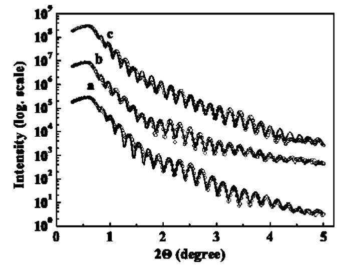

FIG. 1. Low angle x-ray reflectivity scans for 3 selected samples all having $d_{Fe}=17$ Å from Table I. (a) From the series (2); (b) from series (3), and (c) from series (4). The solid lines show calculated reflectivities using the thicknesses given in Table I and assuming an average interface roughness of 4 Å for all interfaces.

Well resolved oscillations reflecting the total layer thickness are clearly seen. Fits using the modified Parratt formalism[30,31] give an interface roughness of less than 4 Å, indicating a high interface quality of the samples. The fits are presented as solid lines in Fig. 1 and are typical for all samples of the present investigation. Radial Bragg scans reveal the (001) texture of all layers in the samples.

Superconducting quantum interference device (SQUID) magnetization measurements were performed at $T=20$ K. Assuming that the saturation magnetization of the Fe layers does not depend on d_{Fe}, as follows from the results of our study of the V/Fe system,[8] we refined the thickness of the Fe layers. These thickness values are used in the plots of Fig. 4.

III. EXPERIMENTAL RESULTS

The superconducting transition temperature T_c was measured resistively in a standard four-terminal configuration by a conventional dc technique with the leads attached to the sample by silver epoxy. For all samples we determined a residual resistivity ratio $RRR=R(300\text{ K})/R(6\text{ K})$ of about 4 and a residual resistivity at 6 K of $\rho(6\text{ K})\simeq 6\mu\Omega\cdot$cm. Using the Pippard relation[32] we can calculate the mean free path of the conduction electrons $l=40$ Å. From the BCS coherence length $\xi_o=440$ Å for pure V and the dirty limit formula $\xi_s=\sqrt{\xi_o l/3.4}$ we determine a superconducting coherence length of $\xi_s\simeq 75$ Å.

Figure 2 shows examples of the resistively measured superconducting transitions. The width of the transitions typi-

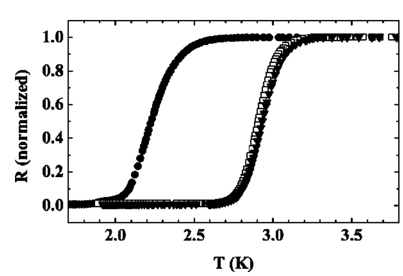

FIG. 2. Superconducting transition measured with dc resistivity for 3 samples, all with fixed $d_{Fe}=17$ Å from Table I. Full circles: from series (2); empty squares: from series (3); and full triangles: from series (4).

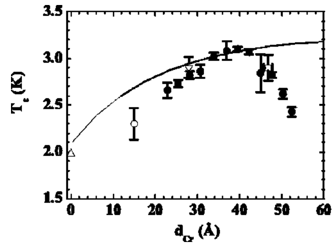

FIG. 3. The superconducting transition temperature as a function of the Cr-layer thickness for all samples from series (1) in Table I. The T_c values for the series (2)–(4) at maximum thickness d_{Fe} are also included [empty symbols corresponding to the symbols used in Figs. 4(b)–4(d)]. The experimental point at $d_{\mathrm{Cr}}=0$ Å has been taken from Ref. 8. The solid line is a theoretical curve (see main text).

cally is less than 0.2 K. The superconducting transition temperature T_c was defined as the midpoint of the transition curve.

In Fig. 3 we plot the T_c values as measured for the samples from series (1) i.e., for the series with a fixed $d_{\mathrm{Fe}}=50$ Å and d_{Cr} varied. In the diagram we have included experimental points (open symbols) from series (2)–(4) taken for the maximum Fe-thickness of about 20 Å (see Table I), and one experimental point for $d_{\mathrm{Cr}}=0$ taken from Ref. 8. Since the penetration depth of the Cooper pairs into Fe is about 10 Å only,[5] the Fe-thickness on the order of 20 Å for the series (2)–(4) is physically equivalent to 50 Å of Fe for the series (1). Consistent with this we find that the additional points in Fig. 3 fit smoothly into the systematics of the series (1) samples. Note that $T_c(d_{\mathrm{Cr}})$ first increases continuously with increasing Cr thickness up to $d_{\mathrm{Cr}}=40$ Å, followed by a sharp drop for thicker Cr-layers.

In series (2)–(4) we have kept d_{Cr} constant at $d_{\mathrm{Cr}}=15$, 28, and 47 Å and varied the thickness of the Fe-layer. The results for the transition temperatures of these three series are reproduced in Figs. 4(b)–4(d) and compared to our previous results on Fe/V/Fe trilayers [Fig. 4(a)]. The solid lines in Fig. 4 show model calculations discussed in the next section. The experimental values for $d_{\mathrm{Fe}}=0$ are taken from our previous experimental investigation of Cr/V/Cr trilayers and corrected for the present *RRR*-values.

The salient features of the results shown in Fig. 4 are as follows: (1) The overall shape of the $T_c(d_{\mathrm{Fe}})$-curve is similar to that obtained for Fe/V/Fe. (2) The amplitude of the initial drop in T_c decreases with increasing thickness of the interleaved Cr-layer. (3) At $d_{\mathrm{Cr}}=47$ Å in Fig. 4(d) the Fe-layers have virtually no influence on T_c any more, indicating that the amplitude of the pair wave function in the Fe-layer is effectively vanishing. These features are obviously due to the screening effect of the Cr-layer, since with increasing d_{Cr} the Cooper pair density reaching the Fe-layer is continuously diminished and the effect of the strong exchange field in Fe is weakened. Thus we estimate a penetration depth of the pair wave function in Cr of about 40 Å, consistent with our previous result on Cr/V/Cr trilayers.[18]

IV. DISCUSSION

First we discuss the model calculations shown by the solid lines in Fig. 3 and in Figs. 4(a)–4(d). Neglecting the

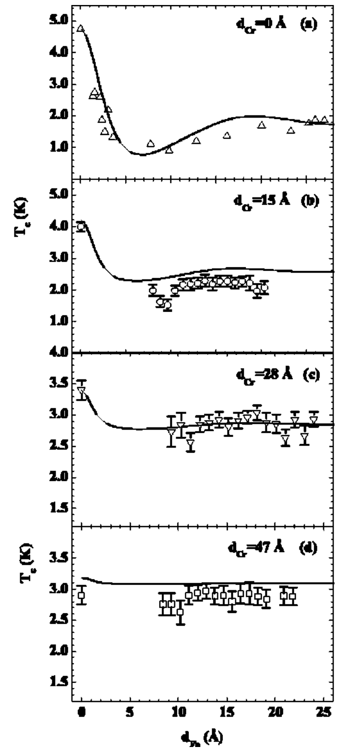

FIG. 4. Superconducting transition temperature as a function of the Fe-layer thickness for samples from series (2)–(4) in Table I. (b) Series (2) with $d_{\mathrm{Cr}}=15$ Å; (c) series (3) with $d_{\mathrm{Cr}}=28$ Å; (d) series (4) with $d_{\mathrm{Cr}}=47$ Å. The corresponding curve for Fe/V/Fe trilayers is taken from Ref. 8 and shown in (a) for comparison. The experimental points at $d_{\mathrm{Fe}}=0$ in (b)–(d) have been taken from Ref. 18. The solid lines are calculations according to a model explained in the main text.

complications caused by the SDW state of antiferromagnetic Cr, we follow the standard procedures described in the literature[10,12–18] and treat the proximity effect of the V/Cr interface by the conventional theory for S/N metal films, originally developed by Werthamer.[33] In addition we allow for a Cooper pair breaking scattering of Abrikosov-Gor'kov-type[34] in the Cr-layer, i.e., we consider the Cr-layer as a paramagnetic (P) layer characterized by a spin-flip scattering time τ_s.

The theory of the proximity effect for a S/P/F layered system has been developed in Ref. 35 and was originally designed for the description of an S/F interface with a paramagnetic interlayer formed due to intermixing at the S/F interface. We can adopt this theory to our present V/Cr/Fe-system by defining Cr as the P-layer.

Without reproducing the theory of Ref. 35 in detail, we note that, aside from band structure parameters principally known from calculations or experiments on the different single layers, the model calculation requires the transparency value T of each interface as input parameters. In order to simplify the fitting procedure we assume that for the transparency parameter (see Ref. 5 for the exact definition) of the Cr/Fe interface we can take the same value of $T_{\mathrm{Cr/Fe}}=1.6$ as derived previously for the V/Fe interface.[8] This assumption is motivated by the similarities of the band structures of V and Cr and the fact that the Fermi momentum mismatch at the interface with a ferromagnet mainly determines the transparency and can at best be eliminated for one spin direction in the ferromagnet, but not simultaneously for the other. For the same reason we approximate the V/Cr interface as ideally transparent, i.e., we assume $T_{V/Cr}$ to be very large. With these assumptions and all other parameters known from our studies of the Fe/V/Fe trilayers,[8] we have fitted simultaneously all data points in Figs. 3 and 4, with τ_s as the only fitting parameter. All measurements can be best described with $\tau_s = 5 \cdot 10^{-13}$ s. The overall shape of the curves is well reproduced, including the penetration depth of about 40 Å for the superconducting pairing wave function in Cr. This remarkably small penetration depth into Cr is thus clearly proven to result from strong inelastic pair breaking scattering in Cr, leading to an exponential damping of the pair wave function amplitude within the Cr-layer.

There is, however, one additional important experimental result, which our model fails to describe, even in qualitative terms. This is the drop of $T_c(d_{Cr})$ for $d_{Cr} > 40$ Å, clearly seen in Fig. 3. We attribute this feature to a transition of the entire Cr-layers from a nonmagnetic state to an incommensurate SDW state at $d_{Cr} \simeq 40$ Å. We believe that the Cr-layer for thicknesses below 40 Å are nonmagnetic, consistent with the Mössbauer experiments by Mibu *et al.*[26,27] mentioned before. However, in their experiment the Cr layer is confined between V layers on both sides, whereas in our case the Cr has an Fe-boundary on one side and a V-layer on the other side. This changes the boundary condition for the confinement of the SDW state and could alter the magnetic properties of the Cr layers for $d_{Cr} < 40$ Å. Therefore, we cannot completely rule out the possibility that a commensurate SDW state might exist for $d_{Cr} < 40$ Å. Nevertheless, in this case we would expect a strong competition between the commensurate SDW state and the superconducting state, leading to a visible T_c-suppression in the Cr-thickness range below 40 Å, which is not seen in the experiment.

The assumption of a strong suppression of the Cooper pair density by the transition of the Cr layer from a nonmagnetic to a SDW state is plausible by the following reason. As mentioned in the Introduction section, BCS-ordering and SDW-ordering in the same region of the Fermi surface can be considered as competing electronic ordering phenomena. In a theoretical approach of this problem (see, e.g., Ref. 29), it was shown that those parts of the Fermi surface where the nesting feature leads to an SDW state, the formation of the BCS-gap is suppressed and the superconducting transition temperature is lowered correspondingly.

V. SUMMARY AND CONCLUSION

We have studied the proximity effect in the thin film system Fe/Cr/V/Cr/Fe, which clearly shows a screening of the ferromagnetic exchange field in Fe by the interleaved Cr-layers. For d_{Cr} larger than 40 Å the Fe-layers practically have no effect on the superconducting transition temperature of V. This provides an upper limit for the Cooper pairs penetrating into the Cr-layer. If Cr would behave as a normal nonsuperconducting metal such as Cu, the penetration depth at low temperature would reach the μm scale. In the Cr layers the drastically reduced penetration depth of the superconducting pair wave function is ascribed to an effective magnetic spin-flip scattering process at defects with local uncompensated magnetic moments.

In addition, we find an anomalous and strong suppression of T_c for a thickness of Cr above 40 Å, i.e. at a thickness larger than the penetration depth of the Cooper pair wave function. For thicknesses above 40 Å, we suggest the onset of an incommensurate spin density wave state in the Cr layer. We argue that the suppression of T_c is due to a transition from the nonmagnetic state of Cr at layer thicknesses below 40 Å to an incommensurate spin density wave state for larger thicknesses. Thus the proximity effect at the V/Cr interface can be used as a sensitive probe for the magnetic state of thin Cr-layers.

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*Electronic address: ilgiz_garifullin@yahoo.com

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