

Figure 4. Resonance fields of components b and c at the frequency 25.94 GHz in the dimer magnet TlCuCl<sub>3</sub> for various temperatures and magnetic field  $\mathbf{H}||b|$ [13].



Figure 5. Energy level diagram of triplet  $\mathbf{k} = 0$  excitations in the dimer magnet TlCuCl<sub>3</sub> [13].

energy level diagram of a triplet of zero-wave-vector excited states in the dimer magnetic TlCuCl<sub>3</sub> is shown in Fig. 5 (from Ref. 13]). Here,  $\Delta$  is the spin gap in the exchange approximation and  $D_0$  and  $E_0$  are the anisotropy parameters of the effective spin Hamiltonian for a spin triplet in a crystal field. We note that in this case, S = 1 excitations arise as collective states in a crystal with spins S = 1/2 at its sites. In the magnetic field  $H_c$  that closes the spin gap for the lower triplet component, the spin-liquid state loses stability, giving way to a magnetic-field-induced antiferromagnetic ordering [14]. The nonlinear dependence of the frequency on the magnetic field signifies the onset of magnetic order and represents a branch of the antiferromagnetic resonance, as discussed in Ref. [13].

To conclude, magnetic resonance experiments have revealed a variety of collective states that are possible for magnetic ions in a singlet matrix of spin-gap crystals: states with the effective spin  $S_{\text{eff}} = 1/2$  at the ends of S = 1 spin chains; excited spin states with the effective spin  $S_{\text{eff}} = 1$  in spin-gap matrices of crystals carrying either spins S = 1 (Haldane systems) or spins S = 1/2 (dimer spin systems); and, finally, hybrid magnetic resonance modes in which nanoscopic clusters and the triplet excitations of a spin-liquid magnet have their spins involved in collective motions.

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## The superconductor/ferromagnet proximity effect and its potential application in spintronics

## I A Garifullin

## 1. Introduction

The so-called proximity effect in superconductor/ferromagnet (S/F) systems — or, in other words, the interplay of superconductivity and ferromagnetism in thin-film heterostructures — has been the subject of intense research over the past ten years (see, e.g., Ref. [1]). In the past few years, interest in the effect has grown dramatically because of its potential uses in spintronics (see, e.g., Refs [2-5]). In multilayer thinfilm systems, a certain combination of F and S layers can be created in which the superconducting transition temperature  $T_{\rm c}$  may be controlled by the orientation of the magnetizations of the F layers relative to one another. The authors [2] first used the S/F proximity effect to theoretically design a spin valve for the superconducting current. In their scheme, denoted as S/F1/N/F2, the magnetizations of two ferromagnetic layers F1 and F2 are isolated from each other by a nonmagnetic metallic layer N, sufficiently thin for the superconducting pair wave function to penetrate from layer S to layer F2. In a theoretical design proposed by Tagirov [3], the superconducting layer is in contact with F layers on either side (F1/S/F2 spin valve). Calculations predict that in both structures, the parallel orientation of the F layer magnetizations provides lower  $T_c$  compared with the antiparallel orientation. In order to enable varying the relative orientation of F layer magnetizations, an antiferromagnetic film is usually deposited on the F2 layer, whose anisotropy fields have the effect of fixing the magnetization of the layer - after

which a small external magnetic field can be used to vary the magnetization direction of F1. There is only one recent report on the realization of Tagirov's design of the superconducting spin valve based on the three-layer system CuNi/Nb/CuNi [4]. The difference in  $T_c$  between the parallel and antiparallel arrangements was found to be about 6.0 mK. For the superconducting spin valve to be more effective, it is desirable that the system F1/S/F2 have a property known as 're-entrant superconductivity.' This phenomenon was first observed by us in Fe/V/Fe films in Ref. [6], where we first observed the complete disappearance of superconductivity as the thickness of the Fe layers increased (in the thickness interval 0.5  $< d_{\rm Fe} < 1$  nm) and then saw it restored for  $d_{\rm Fe} > 1$  nm. Another necessary condition for the Tagirov scheme to be effective is that the thickness  $d_s$  of the S layer be comparable to or less than the superconducting coherence length  $\xi_s$ . A natural explanation is that for Cooper pairs to 'feel' the relative orientation of the F magnetizations, the F layer separation must not be too much greater than the size of the Cooper pair, i.e.,  $\xi_s$ . But our study showed that in a standard three-layer F/S/F system, the ferromagnetic film (even a thin one) is so effective in destroying Cooper pairs that the minimum thickness of the S layer for which superconductivity still exists is of the order of  $3\xi_s$  [6, 7]. It was therefore necessary to somehow secure F/S/F superconductivity at  $d_s \sim \xi_s$ , and one possible way to do this was to place a screening layer between the F and S layers.

This talk presents results on the superconducting proximity effect in the thin-film system Fe/Cr/V/Cr/Fe with chromium layers acting as screens [8, 9]. In addition to new results concerning the magnetic phase transition that occurs in a Cr layer as its thickness Cr is varied, we were also able to determine the upper limit for  $d_{Cr}$  for use in a spin valve. We also made an attempt to realize the superconducting spin valve design proposed in Ref. [2]. Our idea (see Ref. [5]) was to replace the virtual layer N between layers F1 and F2 by a real, nonmagnetic intermediate layer intended to introduce antiferromagnetic exchange coupling between the magnetizations of the ferromagnetic layers [10]. This makes it possible to rotate the magnetization directions of layers F1 and F2 by varying their relative orientation from antiparallel to parallel using an external magnetic field and to measure the resulting shift  $\Delta T_c$ . Instead of a three-layer film F1/N/F2, we used a superlattice Fe/V, with F (Fe) layers strongly antiferromagnetically coupled through V layers [11]. This choice was dictated by technology-related practical considerations [12].

### 2. The proximity effect in the Fe/Cr/V/Fe system

We investigated four series of Fe/Cr/V/Cr/Fe samples. Series 1 was used to measure  $d_{Cr}$  at the fixed value  $d_{Fe} = 5$  nm. In the remaining three series (2–4), the thickness  $d_{Cr}$  was fixed and the variable quantity was  $d_{Fe}$ . In all samples, the thickness of the V layer was 30 nm.

The dependence  $T_c(d_{Cr})$  for series 1 is shown in Fig. 1. It can be seen that for  $d_{Cr} < 4$  nm, the superconducting transition temperature increases as the thickness of the chromium layers increases. On further increase in  $d_{Cr}$ , the superconducting transition temperature passes a maximum and then decreases at a much faster rate than that at which it increased to the maximum.

The  $T_{\rm c}(d_{\rm Fe})$  dependences for the samples of the three series with different fixed thicknesses of the chromium layer



Figure 1. Dependence of  $T_c$  on the chromium layer thickness in series 1 at the fixed value  $d_{Fc} = 5$  nm of the iron layer thickness.

 $(d_{Cr} = 1.5, 2.8, 4.7 \text{ nm})$  are shown in Figs 2b-d. It is seen that they are generally similar to the one we previously obtained [6] for three-layer samples of Fe/V/Fe (Fig. 2a). In these samples, the superconducting transition temperature first sharply decreases and then passes a minimum and saturates as  $d_{Fe}$  is increased. In Fe/Cr/V/Cr/Fe samples, the amplitude of the initial drop in  $T_c$  decreases with increasing the thickness of the Cr layers separating the Fe and V layers. At  $d_{Fe} = 4.7 \text{ nm}$ ,  $T_c$  becomes virtually completely independent of  $d_{Fe}$ , apparently due to the screening effect of the Cr layers. As  $d_{Cr}$  increases, fewer Cooper pairs reach Fe layers, thus decreasing the effect of the exchange field of Fe on the superconductivity of the V layer. From these results, the penetration depth of Cooper pairs into the Cr layer was estimated to be 4.0 nm.

This last conclusion contradicts the results in Fig. 1, which clearly show that starting from 4.0 nm - i.e., from thicknesses exceeding the penetration depth of Cooper pairs into chromium layers — the value of  $T_c$  dramatically decreases with  $d_{Cr}$ . This unambiguously signifies that at  $d_{\rm Cr} \sim 4$  nm, the chromium layers themselves dramatically change their properties because Figs 2c, d show that the superconductivity-destroying effect of the Fe layers on vanadium is already screened out at such a thickness of the Cr layer. We believe that the sharp drop in  $T_{\rm c}(d_{\rm Cr})$  at  $d_{\rm Cr} > 4$  nm occurs because of the transition of Cr layers from a nonmagnetic state to the incommensurate spindensity-wave (SDW) state at  $d_{\rm Cr} \sim 4$ . The conclusion that chromium layers less than 4 nm thick are nonmagnetic is in line with Moessbauer experiments [13]. The following argument seems to justify the assumption that the transition of Cr layers to the SDW state leads to the strong suppression of superconductivity. The SDW state is formed in chromium by band electrons, which can also form a proximity-effectinduced superconducting state there. The theoretical study of the coexistence of SDWs and superconductivity (see, e.g., Ref. 14]) showed that in those parts of the Fermi surface where nesting favors the formation of the SDW state, the chance for the superconducting gap to form is slim, and  $T_{\rm c}$ turns out to be reduced if the SDW transition temperature exceeds the initial value of  $T_c$ . Thus, the appearance of the antiferromagnetic order in Cr and the penetration of Cooper



Figure 2.  $T_c$  versus the thickness of iron layers for the samples of series 2–4. The corresponding dependence for the three-layer system Fe/V/Fe is reproduced from Ref. [6] for comparison.

pairs into the Cr layer may be regarded as two competing antagonistic types of collective electron ordering.

# 3. Superconducting properties of vanadium layers deposited on the antiferromagnetically coupled superlattice $[Fe_2V_{11}]_{20}$

Six samples of MgO(100)/[Fe<sub>2</sub>V<sub>11</sub>]<sub>20</sub>/V( $d_V$ ) were prepared for study. In this structure, two monolayers of iron (Fe<sub>2</sub>) separated by 11 monolayers of V (V<sub>11</sub>) played the roles of the ferromagnetic layers F1 and F2. The superlattice [Fe<sub>2</sub>V<sub>11</sub>]<sub>20</sub>, in which these alternating layers were repeated 20 times, was coated by a sufficiently thick layer of vanadium (with the thickness  $d_V$  from 16 to 30 nm). It is known [11] that the  $V_{11}$  layer establishes the antiferromagnetic exchange coupling between Fe<sub>2</sub> layers.

Magnetization measurements showed that the parallel orientation of magnetizations of various Fe<sub>2</sub> layers in the superlattice  $[Fe_2V_{11}]_{20}$  occurs in the magnetic field 6.0 kOe. The superconducting transitions measured resistively had the width around 0.1 K. As one would expect for thin films in a vortex-free state, no noticeable broadening of the transition widths was observed in large magnetic fields. The upper critical field  $H_{c2}$  was determined from the middle of the transition.

It is well known (see, e.g., Ref. [15]) that for an ordinary three-layer system Fe/V/Fe, in which the thick V layer prevents the spin valve effect, the upper critical field for a magnetic field perpendicular or parallel to the film plane closely follows the theoretical prediction for a 2D thin film [16]. For the perpendicular orientation, the upper critical field is linear in temperature, and in the parallel case the observed dependence is given by

$$H_{c2}^{\rm par} = \frac{\Phi_0}{2\pi\xi(0)} \frac{\sqrt{12}}{d_{\rm s}} \sqrt{1 - \frac{T}{T_{\rm c}}},$$

where  $\Phi_0 = 2 \times 10^{-7} \text{ G cm}^2$  is the magnetic flux quantum,  $\xi(0)$  is the Ginzburg–Landau superconducting coherence length at T = 0 K, and  $d_s$  is the thickness of the superconducting layer. Figure 3 presents the temperature dependences of the square of the upper critical field  $H_{c2}^{par}(T)$  for samples of  $[Fe_2/V_{11}]_{20}/V(d_V)$ . It is seen that the temperature dependence is ideally described by a straight line at fields above 6.0 kOe and increasingly deviates from it below 6.0 kOe. Extrapolating the straight line yields a superconducting transition temperature that is lower than the measured zero-magnetic-field value by more than 0.1 K. A comparison with the magnetization curve of the superlattices  $[Fe_2/V_{11}]_{20}$ shows that the value 6.0 kOe at which the F layer magnetizations turn out to be parallel correlates well with the onset of the linear dependence of  $(H_{c2}^{par}(T))^2$ . This suggest that the deviation in the behavior of the upper critical field from the 2D behavior is due to the gradual change in the relative orientation of the sublattice magnetizations in the superlattice  $[Fe_2/V_{11}]_{20}$  from the parallel orientation in the field above 6.0 kOe to the antiparallel in the zero field. For the sample with  $d_{\rm V} = 16$  nm, the superconducting transition temperature is  $T_c = 1.78$  K, whereas extrapolation from the region of ferromagnetic saturation yields  $T_c = 1.67$  K. Analysis shows that the 0.11 K difference in  $T_c$  is due to the superconducting effect of the spin valve.

#### 4. Conclusion

The study of the proximity effect in a thin-film layered system Fe/Cr/V/Cr/Fe clearly demonstrates the strong screening effects of the Cr layers placed between the superconducting layer of V and the Fe layers that destroy the Cooper pairs. At the chromium layer thickness  $d_{\rm Cr} > 4$  nm, the layers of iron already have practically no effect on  $T_{\rm c}$  for vanadium. From this fact, the upper limit of the penetration depth of Cooper pairs into Cr layers is determined to be 4.0 nm. If the chromium layers behaved as a normal nonsuperconducting metal — for example, Cu — then the penetration depth would reach micrometer values at low temperatures. In chromium



**Figure 3.** The square of the parallel upper critical field as a function of the temperature for the samples  $[Fe_2V_{11}]_{20}/V$  (16 nm) (a) and  $[Fe_2V_{11}]_{20}/V$  (30 nm) (b). Solid line is the linear extrapolation of the temperature dependence from the region of large fields.

layers, the penetration depth of the superconducting pair wave function is determined by electron scattering from defects with an uncompensated local magnetic moment. This effect — the strong screening by chromium layers of the exchange field created by the F layer — will hopefully be used in our further attempts to realize the Tagirov spin valve design.

We have also examined the superconducting spin valve effect in a layer of V deposited on an antiferromagnetically coupled lattice [Fe/V]. Our experiments showed that the superconducting transition temperature of a vanadium film is very sensitive to the relative orientation of the Fe<sub>2</sub> layers of the antiferromagnetically coupled superlattice [Fe<sub>2</sub>/V<sub>11</sub>]<sub>20</sub>. Clearly, F layers in our system cannot be switched easily from the antiparallel to the parallel state because this transition occurs gradually as the external magnetic field is varied from 0 to 6.0 kOe. Still, we hope that it is possible to build a switching devise by replacing the antiferromagnetically coupled superlattice [Fe<sub>2</sub>/V<sub>11</sub>]<sub>20</sub>.

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