Solid State Phenomena Vol. 190 (2012) pp 409-412
Online available since 2012/Jun/14 at www.scientific.net
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doi:10.4028/www.scientific.net/SSP.190.409

Superconducting Critical Temperature and Magnetic Inhomogeneities in Superconductor/Ferromagnet/Superconductor Trilayers

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Keywords: Superconductivity, ferromagnetism, thin films, magnetic nanoclusters, pi-phase.

Abstract. The effect of ferromagnetic layer inhomogeneity on the superconducting resistive transition in Superconductor/Ferromagnet/Superconductor (S/F/S) trilayers is studied. The critical temperature, T_c, and the resistive transition shape versus the F layer thickness, d_F, in Nb/$Cu_{0.41}Ni_{0.59}$/Nb and Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers were analyzed. It is shown that the $T_c(d_F)$ dependence is sensitive to the Ni concentration variation along the F layer for thickness of d_F corresponding to the π-superconducting state and to the 0-π crossover thickness.

Introduction

Superconductivity in structures with alternate Superconducting (S) and Ferromagnetic (F) layers is determined by the proximity effect [1]. The density of Cooper pairs quickly decays in the F-layer due to the exchange field which also causes a nonzero momentum of Cooper pairs creating spatial oscillations of the superconducting pair function. These oscillations manifest themselves in two superconducting critical states of S/F/S trilayers, a “0-state”, with critical temperature T_{c0}, and a “π-state”, with critical temperature $T_{c\pi}$, depending on the F layer thickness d_F. As a result a non-monotonic dependence of the critical temperature T_c versus d_F appears [1]. The oscillation of the superconducting order parameter in S/F system is governed by the coherence length in the ferromagnet, ξ_F, which in turn, is related to the exchange energy, $\xi_F = (\hbar D_F/E_{ex})^{1/2}$ (D_F being the diffusion coefficient of F). This implies that the $T_c(d_F)$ dependence is sensitive to changes in E_{ex}. Such variations can be present in one sample due to inhomogeneity of the ferromagnetic layer.

In the study of properties of S/F proximity coupled hybrids, weakly ferromagnetic alloys, like $Cu_{1-x}Ni_x$ [2] and $Pd_{1-x}Ni_x$ [3], are of great importance. In these systems E_{ex} is controlled by changing the amount of the magnetic element in the alloy. In $Pd_{1-x}Ni_x$ the magnetism is established at much lower Ni percentages compared to $Cu_{1-x}Ni_x$. In fact, because Pd is a highly paramagnetic material [4], the Ni critical concentration x_c, which corresponds to the appearance of the ferromagnetic ordering in $Pd_{1-x}Ni_x$ alloys, is very small, i.e. $x_c \approx 0.02$ [5]. On the other hand, the ferromagnetic order appears in $Cu_{1-x}Ni_x$ alloys at $x_c \approx 0.43$ [6]. Due to this difference in x_c the induced structural disorder is expected to be lower, and consequently, the magnetic ordering to be more homogeneous in $Pd_{1-x}Ni_x$ than in $Cu_{1-x}Ni_x$. Indeed, in $Cu_{1-x}Ni_x$ films for $x > 0.4$ Ni-rich areas tend to form, with typical dimensions of 10 nm [7]. In $Pd_{1-x}Ni_x$ films the Ni segregation is much smaller, and the nanoclusters have smaller dimensions, typically around 3 nm [8], but they are still ferromagnetic [9].

In this contribution we study the influence of E_{ex} variation within the F layer on the superconducting properties of Nb/$Cu_{0.41}Ni_{0.59}$/Nb and Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers with different values of d_F.

Experimental Results

Sample Preparation and Characterization. Nb/$Cu_{0.41}Ni_{0.59}$/Nb and Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers were grown on Si(100) substrates in a UHV *dc* diode magnetron sputtering system. For each of the two systems a complete series of samples has been grown: in both the series the superconducting Nb layers have constant thickness, d_{Nb} = 14 nm, while $Cu_{0.41}Ni_{0.59}$ layer thickness, d_{CuNi}, was varied in the range from 0 to 15 nm and $Pd_{0.81}Ni_{0.19}$, d_{PdNi}, from 0 to 9 nm. The detailed description of the fabrication procedure was published elsewhere [10]. The Ni concentration in the alloys (59 % for CuNi and 19 % for PdNi) was checked by Rutherford Back Scattering (RBS) analysis. The high quality layering of our samples was confirmed by X-ray reflectivity measurements. The roughness in both sets of S/F/S trilayers was comparable and did not exceed 0.8 nm. Finally, the resistive superconducting transitions $R(T)$ were measured in a ^{4}He cryostat using a standard *dc* four-probe technique on unstructured samples typically (10 × 2) mm^2.

Measurements. The $T_c(d_F)$ dependence for Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers is shown in Fig. 1. Here T_c was defined as the temperature where $R = 0.1\ R_N$, (open squares) and $R = 0.9\ R_N$ (closed circles), with R_N the resistance at $T = 10$ K. In Fig. 1 are also shown the theoretical dependencies for T_c versus d_F calculated, in the framework of the Usadel formalism [11, 12], for the system in the 0-state ($T_{c0}(d_{CuNi})$, black solid curve) and in the π-state ($T_{c\pi}(d_{CuNi})$, black dashed curve). In the theoretical simulation the following parameters have been used: the low temperature resistivity $\rho_{CuNi} = 60\ \mu\Omega\times$cm, the exchange energy $E_{ex}^{CuNi} = 140$ K and the diffusion coefficient $D_{CuNi} = 5.3\times10^{-4}$ m^2/s [13]. From that we get $\xi_{CuNi} = 5.4$ nm. The critical temperature of bulk Nb was $T_S = 8.6$ K, which implies that the characteristic length of the diffusive motion of Cooper pairs in the ferromagnet is $\xi_{CuNi}^* = (\hbar D_{CuNi}/2\pi k_B T_S)^{1/2} = 8.5$ nm. Taking for the Nb resistivity $\rho_S = 17\ \mu\Omega\times$cm [10], we get $p \equiv \rho_S/\rho_F = 0.28$. So the only free fit parameter is $\gamma_B \equiv (R_B/\rho_F\xi_F^*)$, where R_B is the S/F interface resistance times its area. γ_B describes the effect of the S/F interface transparency [14]. Fitting the $T_c(R = 0.1\ R_N)$ data reported in Fig. 1 we obtained $\gamma_B = 0.3$, in agreement with results reported for Nb/$Cu_{0.41}Ni_{0.59}$ system [14]. Apart from this standard behavior of $T_c(d_{CuNi})$ we note that some data spread is present in the thickness range 2.5 nm < d_{CuNi} <8 nm. Moreover, as shown in Fig. 1, in this thickness range the width of the transition curves, ΔT_c, defined as $\Delta T_c \equiv T(R = 0.9R_N) - T(R = 0.1R_N)$, increases strongly, reached the value of 0.6 K, while outside this range the transition curves are sharp ($\Delta T_c \approx 0.1$ K). The observed broadening can be due to interface roughness or, in general, to in-plane non-homogeneity of the material which generates a network of Josephson 0- and π-contacts with a subsequent spontaneous nucleation of vortices [15]. The theoretical results (solid and dashed grey curves) presented in Fig. 1, which take into account the magnetic non-homogeneity of F layer, will be discussed later.

In Fig. 2 T_c as a function of d_F for Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers is reported. For this system ΔT_c was always less than 0.1 K and the points corresponding to the critical temperature $T_c \equiv T(R = 0.1R_N)$ (open squares) and to $T_c \equiv T(R = 0.9R_N)$ (closed circles) practically coincide. The solid and the dashed black lines are, respectively, the $T_{c0}(d_{PdNi})$ and the $T_{c\pi}(d_{PdNi})$ dependencies obtained using the following parameters: $\rho_{PdNi} = 64\ \mu\Omega\times$cm, $E_{ex}^{PdNi} = 230$ K, $D_{PdNi} = 2.3\times10^{-4}$m^2/s [16]. As a result, we obtained $\xi_{PdNi} = 2.8$ nm and, using $T_S = 8.3$ K, $\xi_{PdNi}^* = 5.8$ nm. We finally calculated $p = 0.26$ so that the fitting procedure of the experimental data gave $\gamma_B = 0.26$ in reasonable agreement with the results obtained in [16]. The results described by the solid and by the dashed grey lines are discussed in the following section.

Discussion

The main result following from the experimental data reported in Figs. 1 and 2 is that the width of the resistive transition of Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers is much smaller than the one measured for Nb/$Cu_{0.41}Ni_{0.59}$/Nb. However, as noted above, the interface roughness in both systems is typically around 0.8 nm, while Ni clustering is more pronounced in CuNi. In these clusters the value of the exchange energy, E_{ex}^{cl}, is much greater than its value in outside them. Since the lateral dimensions

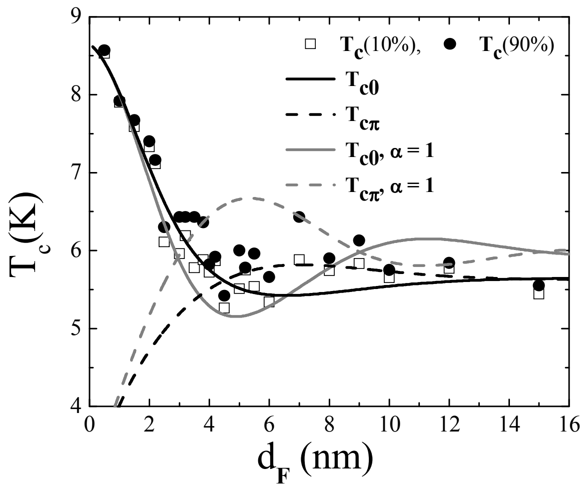

Figure 1. T_c versus d_{CuNi} of Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers. The solid (dashed) black line corresponds to the $T_{c0}(d_{CuNi})$ ($T_{c\pi}(d_{CuNi})$) dependence obtained using the parameters quoted in the text. The solid (dashed) grey line corresponds to the $T_{c0}(d_{CuNi})$ ($T_{c\pi}(d_{CuNi})$) dependencies obtained using the Tagirov correction [17] in the Usadel formalism [12]. See the text for further details.

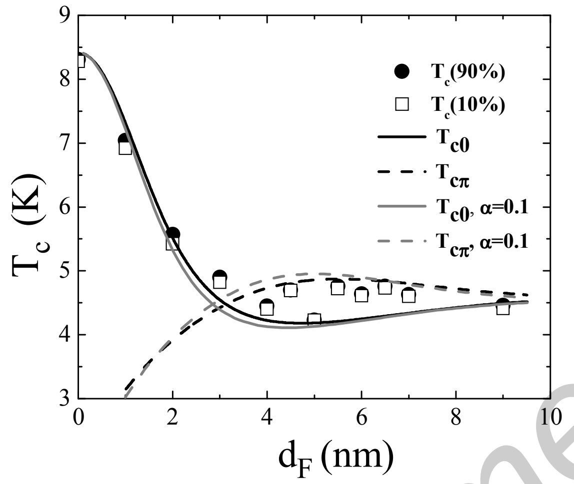

Figure 2. T_c versus d_{PdNi} of Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers. Open squares (closed circles) represent $T_c \equiv T(R=0.1R_N)$ ($T_c \equiv T(R=0.9R_N)$). The solid (dashed) black line corresponds to the $T_{c0}(d_{PdNi})$ ($T_{c\pi}(d_{PdNi})$) dependencies obtained using the parameters quoted in the text. The solid (dashed) grey line corresponds to the $T_{c0}(d_{PdNi})$ ($T_{c\pi}(d_{PdNi})$) dependencies obtained using the Tagirov correction [17] in the Usadel formalism [12]. See the text for further details.

of these clusters are greater than the film thickness, in some points of the S/F/S structure the S-layers will be connected through a stronger ferromagnet, forming S/F^{cl}/S contacts which will be described by different microscopic parameters. In particular, these contacts will be characterized by a different T_c^{cl} versus d_F curve. In the following we will estimate the $T_c^{cl}(d_F)$ dependence applying the Tagirov theory [17]. It has been shown [17, 18] that the diffusive limit of the microscopic theory is not completely adequate when considering S/F structures if F is a strong ferromagnet. It has been proposed that, in order to describe the superconducting properties of S/F structures with a ferromagnet for which $l_F \sim \xi_F$, it is necessary to take into account the first correction to the equations which describe the system in the diffusive limit [17-19]. This leads to a renormalization of the diffusive coefficient, i.e. $D_F \rightarrow D_F^{\pm}=D_F/(1 \pm i\alpha \mathrm{sgn}\omega)$ in the Usadel equations, which describe the superconducting condensate in a ferromagnet. Here $\alpha \equiv l_F/5\xi_{F,m}$, where $\xi_{F,m} \equiv \hbar v_F/2E_{ex}$ is the magnetic stiffness length. The results of the calculations with the renormalized diffusion coefficient, are reported in Fig. 1 for $\alpha = 1$. The range $0 < \alpha < 1$ reproduces well the T_c spread of the experimental data. This result, in our opinion, is related to the presence of relatively large Ni clusters in the $Cu_{0.41}Ni_{0.59}$ alloy. These clusters act as a strong ferromagnet, making the Tagirov arguments applicable to our systems. From the α value one can roughly estimate the E_{ex}^{cl}, the value of the exchange energy in the Ni clusters since, $E_{ex}^{cl} \approx (5\hbar v_F \alpha)/[k_B(2l_{Ni})]$. Choosing for Ni $v_F = 0.28\times10^6$ m/s [20] and $l_{Ni} \approx 2$ nm [21] and using $\alpha = 1$ we find that E_{ex}^{cl} can reach the value of 2.6×10^3 K, which is reasonable value for elemental Ni [20]. From the experiment it also follows that, for $d_{CuNi} \geq 10$ nm, the $R(T)$ dependencies become sharp again. This fact supports our assumption that in some part of the CuNi layers the Ni clusters can form the S/F^{cl}/S contacts. When d_{CuNi} exceeds the average dimension of the cluster this does not considerably affect the superconducting properties of the S/F/S structure. Finally in Fig. 2 we report the results for the Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayers obtained by applying the same procedure followed above for the

$Nb/Cu_{0.41}Ni_{0.59}/Nb$ trilayers. Black lines refer to α = 0 and grey lines to α = 0.1. The agreement with the experimental data is worse in the last case. We believe that this result is a direct consequence of the smaller dimension of the Ni clusters in $Pd_{0.81}Ni_{0.19}$. For this reason the behavior of $T_c(d_{PdNi})$ can be satisfactory described by the standard method.

Summary

A systematic study of the $T_c(d_F)$ dependence in S/F/S trilayers, with F being a weakly ferromagnetic alloy, has been performed. The experimental data have been analyzed by applying the approach developed by Tagirov to describe superconducting/strong ferromagnetic systems. In the present case the aim was to take into account the possible presence of Ni segregation in the alloys. The model successfully reproduces the data for the $Nb/Cu_{0.41}Ni_{0.59}/Nb$ system, while it is evidently not suited for the $Nb/Pd_{0.81}Ni_{0.19}/Nb$ data. We ascribe this result to the different properties of the weak ferromagnetic alloys, namely to a more pronounced clustering in the $Cu_{0.41}Ni_{0.59}$ case. Finally, the indirect quantitative estimate of the exchange energy in the clusters supports our argument.

We are grateful to L.R. Tagirov for valuable discussions. The work is partially supported by the BFBR, grant F10R-063 (V.N.K. and S.L.P.), RFBR, grant 10-02-90014 (M.Yu.K.) and by the Italian MIUR-PRIN 2007 project *Proprietà di trasporto elettrico dc e ac di strutture ibride stratificate superconduttore/ferromagnete realizzate con materiali tradizionali (C.C. and C.A.).*

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10.4028/www.scientific.net/SSP.190

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10.4028/www.scientific.net/SSP.190.409