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# Effect of the variation of the exchange energy on the superconducting critical temperature of S/F/S trilayers

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**Abstract.** The effect of the exchange energy variation in weakly ferromagnetic alloys on the superconducting resistive transition of superconductor/ferromagnet/superconductor (S/F/S) trilayers is studied. Critical temperature, $T_c$, and resistive transitions versus the F-layer thickness, $d_F$, have been analyzed in $Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers. We show that $T_c(d_F)$ dependence is sensitive to magnetic inhomogeneities in the F-layer for values of $d_F$ corresponding to thickness range where the $\pi$-superconducting state is established.

## 1 Introduction

Superconductivity in structures with alternated 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 a spatial oscillation of the superconducting pair function [2]. These oscillations manifest themselves in two superconducting critical states of S/F/S trilayers, a "0-state", with critical temperature $T_{c0}$, and a "$\pi$-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 $d_F$, the F layer thickness, appears [3,4].

The existence of the $\pi$-state and other non-trivial properties of S/F systems, make these structures very appealing, being good candidates in the field of spintronics and digital electronics [1,5,6]. For these reasons, these promising systems deserve deep investigation, also in regard fundamental problems such as, for instance, interface quality or the magnetic properties of the ferromagnetic layer. The oscillation of the superconducting order parameter in S/F systems is governed by the coherence length in the ferromagnet, $\xi_F$. If the ferromagnet is in the diffusive regime and $E_{ex} \gg k_B T$, $\xi_F = \sqrt{\hbar D_F / E_{ex}} = \sqrt{\hbar v_F \ell_F / 3E_{ex}}$ (here $D_F$, $v_F$, $E_{ex}$ and $\ell_F$ are the diffusion coefficient, the Fermi velocity, the exchange energy and the electron mean free path of the ferromagnet, respectively). This implies that the $T_c(d_F)$ dependence is sensitive, especially for $d_F$ values close to the thickness value where the 0-$\pi$ crossover takes place, 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 $Cu_{1-x}Ni_x$ [7,8] and $Pd_{1-x}Ni_x$ [9], are of great importance. In these systems $E_{ex}$ is controlled by changing the amount of the magnetic element in the alloy, and the superconducting order parameter can be induced in the F-layer over distances up to about ten nanometers [10]. 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 [11], 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, $x_c \approx 0.02$ [12–14]. On the other hand, Cu is a diamagnetic material and therefore the ferromagnetic order appears in $Cu_{1-x}Ni_x$ alloys at much larger values of the Ni concentration, namely at $x_c \approx 0.43$ [15]. 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 $Cu_{1-x}Ni_x$ [16]. Indeed, in $Cu_{1-x}Ni_x$ alloys for $x > 0.4$ Ni-rich areas tend to form, with typical dimensions of 10 nanometers. The presence of such Ni nanoclusters has been detected in $Cu_{1-x}Ni_x$ both in bulk materials [17,18] and in thin films [19–21]. In $Pd_{1-x}Ni_x$ films the Ni segregation is much smaller, and the nanoclusters have smaller dimensions, typically around 3 nanometers [20], but they are still ferromagnetic [22].

In this paper 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

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with different values of $d_F$. We show that the presence of local disturbances of the magnetic homogeneity are responsible for the broadening of the zero magnetic field superconducting transition curves for $Nb/Cu_{0.41}Ni_{0.59}/Nb$ samples observed in the thickness range where the $\pi$-state takes place.

## 2 Experimental

$Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers were grown on Si(100) substrates in a UHV dc diode magnetron sputtering system with a base pressure less than $10^{-9}$ mbar and a sputtering Argon pressure of $4 \times 10^{-3}$ mbar. 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, $d_{\rm Nb} = 14$ nm, while $Cu_{0.41}Ni_{0.59}$ layer thickness, $d_{\rm CuNi}$, was varied in the range from 0 to 15 nm and $Pd_{0.81}Ni_{0.19}$, $d_{\rm PdNi}$, from 0 to 9 nm. To prevent Nb oxidation a 2 nm thick Al cap layer was finally deposited on the top of the structures. The samples' fabrication was performed under identical conditions. Six substrates were transferred one at a time from the load-lock to the deposition chamber. Samples were always positioned exactly in the center of the main chamber to achieve very similar deposition conditions for all the samples. The detailed description of the fabrication procedure was published elsewhere [23].

The Ni concentration in the alloys (59% for CuNi and 19% for PdNi) was checked by Rutherford BackScattering (RBS) analysis. The Curie temperature, $T_{Curie}$, and the magnetic moment per atoms, $\mu_{at}$ at $T = 10$ K, were estimated to be $T_{Curie} \approx 220$ K and $\mu_{at} \approx 0.12\ \mu_B/at$ for $Cu_{0.41}Ni_{0.59}$ [24], and $T_{Curie} \approx 210$ K and $\mu_{at} \approx 0.3\ \mu_B/at$ for $Pd_{0.81}Ni_{0.19}$ [25]. The high quality layering of our samples was confirmed by X-ray reflectivity measurements. The obtained data were fitted using the Parrat and Nevot-Croce recursion relation, which takes into account the electron density height fluctuations at the interface [26,27]. The fit gives information about the presence of interface roughness at different interfaces. We obtained that the roughness in both sets of S/F/S trilayers was comparable and did not exceed 0.8 nm [28,29]. 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 $(10 \times 2)$ mm$^2$.

## 3 Results

The superconduting critical temperature, $T_c$, as a function of $d_F$ for $Nb/Cu_{0.41}Ni_{0.59}/Nb$ trilayers is shown in Figure 1. In this case $T_c$ was defined as the temperature where $R = 0.1\ R_N$, with $R_N$ the resistance at $T = 10$ K. In Figure 1 are also shown the theoretical dependencies for $T_c$ versus $d_F$ calculated, in the framework of the Usadel formalism [30] applying the method of the data analysis described in [31], for the system in the 0-state ($T_{c0}(d_{\rm CuNi})$, solid curve) and in the $\pi$-state ($T_{c\pi}(d_{\rm CuNi})$, dashed curve). In the theoretical simulation the following parameters which enter the model [31] have been used: the low temperature resistivity $\rho_{\rm CuNi} = 60\ \mu\Omega$ cm, the exchange energy $E_{ex}^{\rm CuNi} = 140$ K and the diffusion coefficient $D_{\rm CuNi} = 5.3 \times 10^{-4}$ m$^2$/s [32]. From that, using the expression for $\xi_F$ reported above, we get $\xi_{\rm CuNi} = 5.4$ nm. The critical temperature of bulk Nb was $T_S = 8.6$ K, which implies that the characteristic length of the diffusive motion of the Cooper pairs in the ferromagnet is $\xi^*_{\rm CuNi} = \sqrt{\hbar D_{\rm CuNi}/2\pi k_B T_S} = 8.5$ nm. Taking for the $Nb$ resistivity $\rho_S = 17\ \mu\Omega$ cm [25], we get $p \equiv \rho_S/\rho_F = 0.28$. So the only free fit parameter which is left in the model [31] is $\gamma_b \equiv (R_B/\rho_F\xi^*_F)$, where $R_B$ is the S/F interface resistance times its area. $\gamma_b$ describes the effect of the S/F interface transparency and varies from 0 (ideal interface) to $\infty$ (completely reflective interface) [33]. Fitting the $Nb/Cu_{0.41}Ni_{0.59}/Nb$ data reported in Figure 1 we obtained $\gamma_b = 0.3$, in agreement with results reported in the literature for $Nb/Cu_{0.43}Ni_{0.57}$ systems [33,34]. From Figure 1 it also follows that the crossover between the 0- and the $\pi$-state occurs at $d^*_{\rm CuNi} \approx 4.5$ nm.

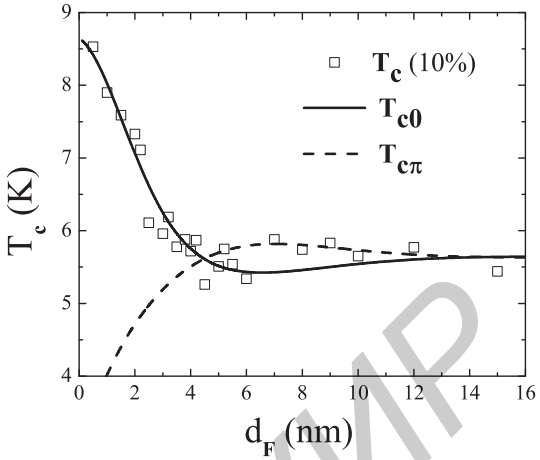


**Fig. 1.** $T_c$ versus $d_{\rm CuNi}$ of $Nb/Cu_{0.41}Ni_{0.59}/Nb$ trilayers with $d_{\rm Nb} = 14$ nm. $T_c$ was obtained from resistive transitions and defined as the temperature where $R = 0.1R_N$. The solid (dashed) line corresponds to the $T_{c0}(d_{\rm CuNi})$ ($T_{c\pi}(d_{\rm CuNi})$) dependence [31] obtained using the parameters quoted in the text.

In Figure 2, in addition to the data reported in Figure 1 (open squares), are also plotted the critical temperature values taken at the onset of the resistive transition (closed circles). $T_c$ is now defined as the temperature where $R = 0.9R_N$. From the result of Figure 2 it follows that at $d_{\rm CuNi}(\approx 2\ {\rm nm}) \ll d^*_{\rm CuNi}$ and for $d_{\rm CuNi}(\approx 10\ {\rm nm}) \gg d^*_{\rm CuNi}$ the resistive transitions are sharp. The width of the transitions, $\Delta T_c \equiv T(R = 0.9R_N) - T(R = 0.1R_N))$, in this case does not exceed 0.1 K, while, close to $d^*_{\rm CuNi}$, $\Delta T_c$ increases up to 0.6 K. Such broadening in the region of the 0-$\pi$ crossover have already been observed in S/F multilayers [35–38] but, to our knowledge, it was not investigated in detail. In reference [39] a qualitative model was developed to explain such effect. In particular, it was proposed that the broadening can be due to in-plane inhomogeneity

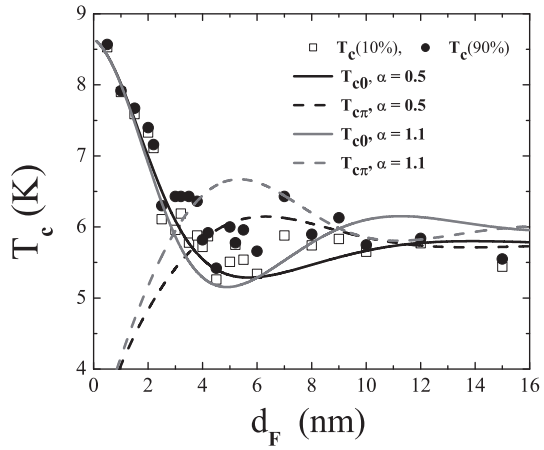


**Fig. 2.** $T_c$ versus $d_{\mathrm{CuNi}}$ of $\mathrm{Nb/Cu_{0.41}Ni_{0.59}/Nb}$ trilayers with $d_{\mathrm{Nb}} = 14$ nm. Open squares (closed circles) represent $T_c \equiv T(R = 0.1R_N)$ ($T_c \equiv T(R = 0.9R_N)$). The solid (dashed) line corresponds to the $T_{c0}(d_{\mathrm{CuNi}})$ ($T_{c\pi}(d_{\mathrm{CuNi}})$) dependencies obtained using the Tagirov correction [41] in the Usadel formalism [31]. See the text for further details.

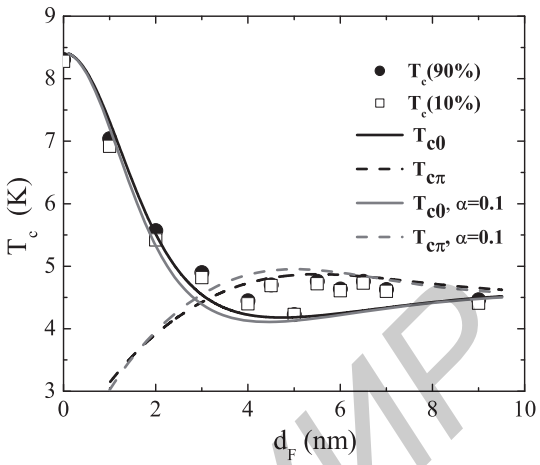


**Fig. 3.** $T_c$ versus $d_{\mathrm{PdNi}}$ of $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ trilayers with $d_{\mathrm{Nb}} = 14$ nm. Open squares (closed circles) represent $T_c \equiv T(R = 0.1R_N)$ ($T_c \equiv T(R = 0.9R_N)$). The solid (dashed) black line corresponds to the $T_{c0}(d_{\mathrm{PdNi}})$ ($T_{c\pi}(d_{\mathrm{PdNi}})$) dependencies [31] obtained using the parameters quoted in the text. The solid (dashed) grey line corresponds to the $T_{c0}(d_{\mathrm{PdNi}})$ ($T_{c\pi}(d_{\mathrm{PdNi}})$) dependencies obtained using the Tagirov correction [41] in the Usadel formalism [31]. See the text for further details.

of the materials which generates a network of Josephson 0- and $\pi$-contacts with a subsequent spontaneous nucleation of vortices [39]. In this paper we analyze quantitatively this effect paying the main attention to the possible influence of the magnetic inhomogeneity of F-layer on the broadening. Since the interface transparency is comparable for the two systems we do not consider it as a possible cause of the enlargement of the resistive transitions. The theoretical results (solid and dashed curves) presented in Figure 2 will be discussed later.

In Figure 3 $T_c$ as a function of $d_F$ for $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ trilayers is reported. For this system the width of the resistive transitions, $\Delta T_c$, was always less than 0.1 K even for thickness of the F-layer close to $d^*_{\mathrm{PdNi}}$ (which, in this case, is 3.1 nm) 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 $T_{c0}(d_{\mathrm{PdNi}})$ and the $T_{c\pi}(d_{\mathrm{PdNi}})$ dependencies obtained using the model of reference [31] in which the following parameters have been used: $\rho_{\mathrm{PdNi}} = 64\ \mu\Omega$ cm, $E_{ex}^{\mathrm{PdNi}} = 230$ K, $D_{\mathrm{PdNi}} = 2.3 \times 10^{-4}$ m$^2$/s [25]. As a result, we obtained $\xi_{\mathrm{PdNi}} = 2.8$ nm and, using $T_S = 8.3$ K, $\xi^*_{\mathrm{PdNi}} = 5.8$ 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 [25]. The small discrepancy between this value and the one reported in [25] for $\mathrm{Nb/Pd_{0.81}Ni_{0.19}}$ bilayers is probably due to the different configurations of the analyzed samples. It has been recently shown that for the Nb/Pd systems the layer deposition sequence significantly affects the structural disorder at the interfaces and, consequently, the critical temperature of the whole sample [40]. This work suggests that it can be not straightforward to compare the results obtained for $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ trilayers with the one reported for $\mathrm{Nb/Pd_{0.81}Ni_{0.19}}$ bilayers. The results described by the solid and the dashed grey lines are discussed in the following section.

## 4 Discussion

The main result following from the experimental data reported in Figures 2 and 3 is that the width of the resistive transition of $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ trilayers is much smaller than the one measured for $\mathrm{Nb/Cu_{0.41}Ni_{0.59}/Nb}$. 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 outside them. Since the lateral dimensions 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 $\mathrm{S}/F^{cl}/\mathrm{S}$ 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 [41,42]. It has been shown [41,42] that the diffusive limit of the microscopic theory is not completely adequate when considering S/F structures if F is a strong ferromagnet. In fact, for such materials the characteristic decay length of the superconducting pair function in the F-layer, $\xi_F$, becomes comparable or even smaller than $\ell_F$ and, consequently, the conditions for applicability of equations in the diffusive limit are not fulfilled. It has been proposed that, in order to describe the superconducting properties of S/F structures with a ferromagnet for which $\ell_F \sim \xi_F$, it is necessary to take into account the first correction to the equations which describe the system in the diffusive

limit [41,43]. This leads to a renormalization of the diffusion coefficient, i.e. $D_F \longrightarrow D_F^{\pm} = D_F/(1 \pm i\alpha \mathrm{sgn}\omega)$ in the Usadel equations, which describe the superconducting condensate in a ferromagnet:

$$\left(|\omega| \pm iE_{ex} - \frac{\hbar D_F^{\pm}}{2}\,\partial_{\mathbf{r}}^2\right) F^{\pm}(\omega;\mathbf{r}) = 0. \qquad (1)$$

Here $\alpha \equiv \ell_F/5\xi_{F,m}$, where $\xi_{F,m} \equiv \hbar \mathrm{v}_F/2E_{ex}$ is the magnetic stiffness length, $\omega = \pi k_B T(2n+1)$, $n = 0, 1, \ldots$ are Matsubara frequencies, $F^{\pm}(\omega;\mathbf{r})$ are the anomalous Green functions in the diffusive limit. Calculations revealed that equations taken in the diffusive limit and with the above implementation adequately describe the experimental data [41,42,44–47]. The results of the calculations, performed using the model of reference [31] but considering the renormalized diffusion coefficient, are reported in Figure 2 to fit the data of $Nb/Cu_{0.41}Ni_{0.59}/Nb$ trilayers for two different values of the coefficient $\alpha$: black lines refer to $\alpha = 0.5$ and grey lines to $\alpha = 1.1$. These two values were chosen from the range $0 < \alpha < 1.1$ which reproduces completely the $T_c$ spread of the experimental data.

In our opinion, the obtained result 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 $\alpha$ values one can roughly estimate the $E_{ex}^{cl}$, the value of the exchange energy in the Ni clusters since, in temperature units, $E_{ex}^{cl} \approx (5\hbar \mathrm{v}_F\alpha)/[k_B(2\ell_{\mathrm{Ni}})]$. Choosing for Ni $\mathrm{v}_F = 0.28 \times 10^6$ m/s [48] and $\ell_{Ni} \approx 2$ nm [49] and using $\alpha = 0.5$ or $\alpha = 1.1$, we find that $E_{ex}^{cl}$ changes from $1.3 \times 10^3$ K to $2.9 \times 10^3$ K, which are reasonable values for elemental Ni [48,50]. Also, from the $T_c(d_{\mathrm{Ni}})$ behavior of Nb/Ni bilayers the value $\xi_{F,m} = 0.88$ nm was obtained [49] from which one gets $E_{ex} = 2.4 \times 10^3$ K. From the experiment it also follows that, for $d_{\mathrm{CuNi}} \geq 10$ nm the $R(T)$ curves 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 $d_{\mathrm{CuNi}}$ exceeds the average dimension of the cluster this does not considerably affect the superconducting properties of the S/F/S structure.

In Figure 3 we show 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 $\alpha = 0$ and grey lines to $\alpha = 0.1$. It is evident that 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 $T_c(d_{\mathrm{PdNi}})$ can be satisfactory described by the standard method without taking into account Tagirov's approach.

## 5 Conclusions

In conclusion, 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. For the $Nb/Cu_{0.41}Ni_{0.59}/Nb$ trilayers a broadening of the $R(T)$ transitions is observed in the $\pi$-phase thickness region, where $\Delta T_c = 0.6$ K. On the other hand for the $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers these transitions are always sharp and $\Delta T_c$ does not exceed 0.1 K. 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 $Nb/Cu_{0.41}Ni_{0.59}$ system, while it is evidently not suited for the $Nb/Pd_{0.81}Ni_{0.19}$ 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 cluster supports our argument.

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