Evaluation of the specific boundary resistance of superconducting/weakly ferromagnetic hybrids by critical temperature measurements

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The specific boundary resistance, R_B, of superconducting (S)/ferromagnetic (F) hybrids is obtained by measuring the dependence of the superconducting critical temperature, T_c, as a function of the thickness of the ferromagnetic layers, d_F, and of the thickness of the superconducting layers, d_S, in S/F bilayers and of the number of S/F bilayers, N_{bil}, for large values of N_{bil}, in F/[S/F]$_{N_{bil}}$ multilayers. We choose Nb for the superconducting material and two weakly ferromagnetic alloys, $Cu_{0.38}Ni_{0.62}$ and $Pd_{0.81}Ni_{0.19}$, for F. Analyzing the experimental results by using a matrix formulation of a theoretical model based on the Usadel equations, we are able to determine the value of R_B which for both the S/F hybrids is of the order of 1 fΩ m^2. © *2011 American Institute of Physics*. [doi:10.1063/1.3664748]

I. INTRODUCTION

The study of the interplay between superconductivity (S) and ferromagnetism (F) combined in layered structures represents nowadays a very active area of investigation.[1] As in the case of superconducting/normal metal (N) hybrids the superconductivity in these systems is governed by the proximity effect.[2] In the case of conventional spin-singlet superconductors, due to the spatial inhomogeneous nature of the superconducting order parameter induced in the F-layer, many new effects have been theoretically predicted and experimentally observed. Among them, the non-monotonic behavior of the superconducting critical temperature T_c as a function of the F layer thickness, d_F, in S/F heterostructures[3–5] or the oscillations of the critical current in S/F/S Josephson junctions[6] are worth mentioning. In view of possible applications of these S/F hybrids, what it is obviously very important in order to obtain a good contact between the two metals present in the hybrid structures is the quality of the interface. The parameter which has been added to the theory of the proximity effect to describe the strength of the interactions between two metals is the interface transparency coefficient, $\mathcal{T}$, whose role has been extensively studied in the S/F,[7–17] as well as in the S/N case.[18–24] The role of the interface transparency in the theory is explicitly taken into account introducing the parameter $\gamma_b \equiv R_B/(\rho_F \xi_F)$.[25] γ_b describes the quality of the interface barrier, through the relation $\gamma_b = 2\ell_F/(3t_F\xi_F)$.[9,26] The quantity t_F is the so-called transparency parameter and can vary in the range $[0, \infty]$. $t_F = 0$ means that the system is characterized by a negligible transparency while $t_F = \infty$ means a perfect interface. t_F is, in turn, related to the quantum mechanical interface transparency coefficient, $\mathcal{T}$, through the relation $\mathcal{T} = t_F/(1 + t_F)$.[9,26] In the above equations R_B is the specific interface resistance, ρ_F is the low-temperature resistivity of the F-layer, $\xi_F = \sqrt{\hbar D_F/2\pi k_B T_S}$ (where D_F is the diffusion coefficient in the F-layer and T_S is the critical temperature of the bulk superconductor) is a measure of the diffusive motion of the Cooper pairs in the ferromagnet, ℓ_F is the low-temperature mean free path of the electrons in the ferromagnet. A new experimental method has been recently developed to evaluate the parameter γ_b for S/N hybrids.[24] By simply measuring the dependence of T_c on the Nb layer thickness, d_S, in Cu/Nb/Cu trilayers and on the number of bilayers, N_{bil}, in Cu/[Nb/Cu]$_{N_{bil}}$ multilayers a value of 0.33 fΩ m^2 has been obtained for the specific resistance of the Nb/Cu interface, number which is in very good agreement with the value obtained using a more sophisticated technique based on current perpendicular-to-plane (CPP) measurements.[27]

In this paper we extend the previous method to the more complex case of S/F hybrids. The experimental data, obtained by measuring resistive transition curves on unstructured $Nb/Cu_{0.38}Ni_{0.62}$ hybrids, have been interpreted in the framework of a theoretical model[28] based on the linearized Usadel equations valid for the S/F case, using the Kupriyanov and Lukichev (KL) boundary conditions.[25] We show that different pairs (ξ_F, γ_b) reproduce with the same accuracy the behavior of T_c as a function of d_S for the bilayers. The degeneracy is removed if the asymptotic behavior of the T_c versus N_{bil} in $Cu_{0.38}Ni_{0.62}/[Nb/Cu_{0.38}Ni_{0.62}]_{N_{bil}}$ multilayers is considered. This allows us to extract the value for the specific resistance R_B of the $Nb/Cu_{0.38}Ni_{0.62}$ hybrids. The same approach has been used to evaluate R_B also for the $Nb/Pd_{0.81}Ni_{0.19}$ system.

II. EXPERIMENTAL RESULTS AND DISCUSSION

$Nb/Cu_{0.38}Ni_{0.62}$ multilayers have been grown on Si(100) substrates by UHV dc diode magnetron sputtering in an Ar pressure of 1×10^{-3} mbar after obtaining a base pressure of 2×10^{-8} mbar. The typical deposition rates were 0.40 nm/s for Nb and 0.21 nm/s for $Cu_{0.38}Ni_{0.62}$ as measured by a quartz crystal monitor previously calibrated by low-angle x-ray reflectivity measurements.

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Three sets of samples have been fabricated. The first series of $Nb/Cu_{0.38}Ni_{0.62}$ bilayers was obtained by keeping constant the thickness of the Nb layer to 15 nm and varying the thickness of the CuNi layer in the range 2–10 nm. This series was used to study the T_c versus d_F dependence. The second series of $Nb/Cu_{0.38}Ni_{0.62}$ bilayers was obtained by keeping constant the thickness of the CuNi layer to 15 nm and varying the thickness of the Nb layer in the range 20–100 nm. This series was used to study the T_c versus d_S dependence. The third series consisting of $Cu_{0.38}Ni_{0.62}/[Nb/Cu_{0.38}Ni_{0.62}]_{N_{bil}}$ multilayers with N_{bil} in the range 7–14 was used to study the T_c versus N_{bil} dependence. In this series the thickness of the superconducting and the ferromagnetic layers were, respectively, fixed to $d_S = 15$ nm and $d_F = 5$ nm. All the experimental data have been analyzed in the frame of the theoretical model whose details are described elsewhere.[28] The model is essentially based on the application of the Usadel microscopic equations[29] which can be exactly solved using a matrix method similar to the one used to solve the same problem applied to the case of S/N hybrids.[22] The model contains six independent parameters: T_S, the superconducting coherence length, $\xi_S = \sqrt{\hbar D_S/2\pi k_B T_S}$ (D_S is the diffusion coefficient in the superconducting layer), $p \equiv \rho_S/\rho_F$, the ratio between the low-temperature resistivities of the S- and F-metals, E_{ex}, the exchange energy, ξ_F (or, equivalently, D_F) and, finally, γ_b, the transparency parameter. The value of the superconducting coherence length $\xi_S = 6.2$ nm has been determined by measuring the perpendicular upper critical magnetic field of a single Nb film 15-nm thick through the relation $\xi_S = (2/\pi\sqrt{2\pi})\sqrt{-\Phi_0/[T_c(dH_{c2}/dT)|_{T=T_c}]}$. To estimate E_{ex} we considered that in S/F hybrids the superconducting order parameter does not only decay in the ferromagnetic layer but it also oscillates as a function of the distance from the interface.[30] As a consequence, the superconducting critical temperature shows a non-monotonic behavior as a function of d_F. In Fig. 1 the $T_c(d_F)$ dependence is reported for the first series of $Nb/Cu_{0.38}Ni_{0.62}$ bilayers. Considering that the position of the minimum is strongly determined by the value of E_{ex} (Ref. 10) from $d_F^{min} \approx 6$ nm we obtain $E_{ex} \approx 11$ meV. This number scales well with the values obtained, with different methods, for $Nb/Cu_{1-x}Ni_x$ hybrids for different Ni concentrations of the ferromagnetic alloy.[10,15,31–34] We did not try to make any attempt to fit these data since in this case, due to very small values of d_F, the dependence of ρ_F on d_F should have been explicitly taken into account[35] going beyond the aim of this paper.

Other two undetermined physical quantities present in the model, ξ_F and γ_b, are obtained by fitting the $T_c(d_S)$ dependence of the second series of bilayers, using again the model reported in Ref. 28, as shown in Fig. 2. The line in the figure is, in fact, the theoretical curve obtained using for ξ_S and E_{ex} the values reported above. The value $T_S = 8.3$ K used in the calculations has been fixed by measuring the critical temperature of a 150-nm-thick Nb film deliberately fabricated, while to calculate the value of the parameter p for this series of bilayers ($d_F = 15$ nm) we have used

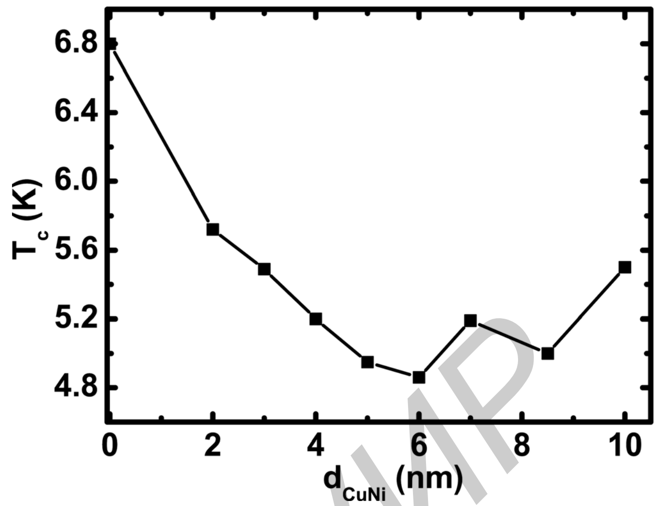

FIG. 1. Superconducting critical temperature, T_c, vs $Cu_{0.38}Ni_{0.62}$ thickness in $Nb/Cu_{0.38}Ni_{0.62}$ bilayers with constant Nb thickness, $d_S = 15$ nm. The line is a guide to the eye.

$\rho_F = 60\ \mu\Omega$ cm which is the low-temperature resistivity measured on a 15-nm-thick $Cu_{0.38}Ni_{0.62}$ film. To fix the value of the parameter p we have measured the low-temperature resistivity of the single Nb film 15-nm-thick, which was equal to 23 $\mu\Omega$ cm resulting in $p = 0.38$. In this way only ξ_F and γ_b were left as fitting parameters. What we found was that, keeping fixed the other four parameters, there is an infinite number of pairs (ξ_F, γ_b) which reproduces the experimental data with exactly the same accuracy. In Fig. 2, we report only the curve calculated using $\xi_F = 6.8$ nm and $\gamma_b = 0.15$, while the inset shows the curve which represents all the pairs (ξ_F, γ_b) which give similar theoretical fit of the $T_c(d_S)$ experimental data. Here we wish to underline that a better description of the experimental $T_c(d_S)$ data can be certainly obtained by allowing ξ_S, p, and E_{ex} to change as fitting parameters. However, since they have been determined from independent measurements they

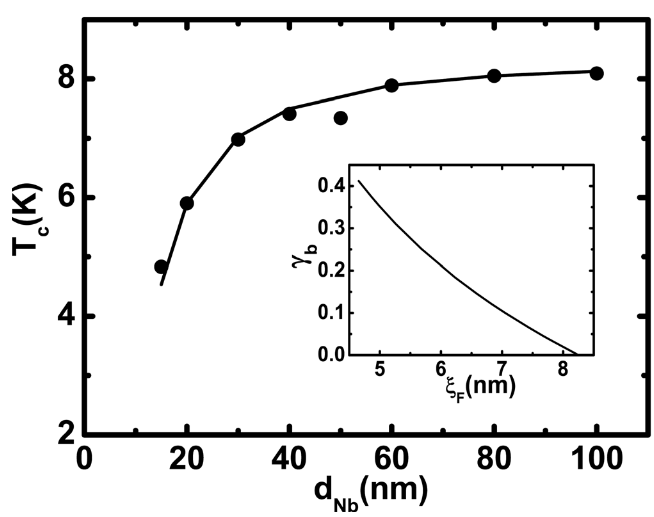

FIG. 2. Dependence of the superconducting critical temperature on the Nb thickness in $Nb/Cu_{0.38}Ni_{0.62}$ bilayers with $d_{CuNi} = 15$ nm. The line is the theoretical fit to the experimental data obtained using $\xi_F = 6.8$ nm and $\gamma_b = 0.15$. Inset: dependence of the parameter γ_b on ξ_F.

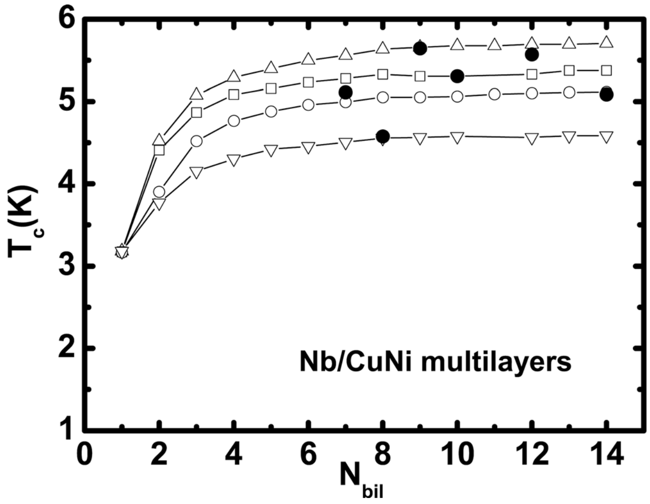

FIG. 3. Dependence of the superconducting critical temperature on N_{bil} in $Cu_{0.38}Ni_{0.62}/[Nb/Cu_{0.38}Ni_{0.62}]_{N_{bil}}$ multilayers with $d_S = 15$ nm and $d_F = 5$ nm (closed circles). Theoretical calculations refer to the following parameters: $\xi_F = 7.3$ nm and $\gamma_b = 0.06$ (up-triangles), $\xi_F = 7.1$ nm and $\gamma_b = 0.09$ (squares), $\xi_F = 6.8$ nm and $\gamma_b = 0.12$ (circles), $\xi_F = 6.4$ nm and $\gamma_b = 0.18$ (down-triangles).

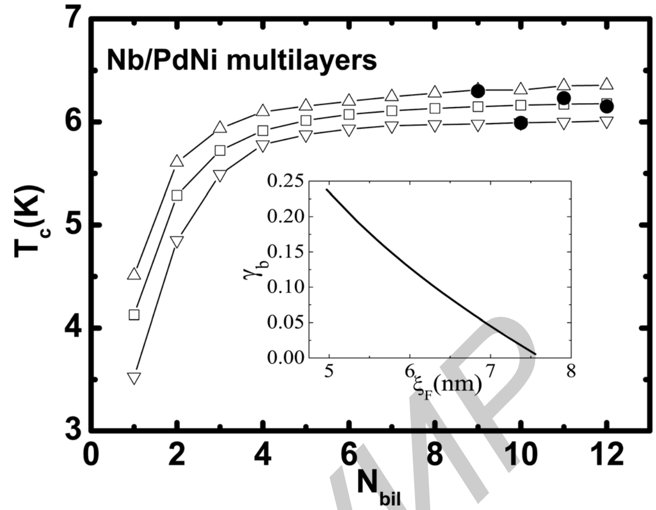

FIG. 4. Dependence of the superconducting critical temperature on N_{bil} in $Pd_{0.81}Ni_{0.19}/[Nb/Pd_{0.81}Ni_{0.19}]_{N_{bil}}$ multilayers with $d_S = 16$ nm and $d_F = 2.2$ nm (closed circles). Theoretical calculations refer to the following parameters: $\xi_F = 5.6$ nm and $\gamma_b = 0.18$ (up-triangles), $\xi_F = 5.4$ nm and $\gamma_b = 0.19$ (squares), $\xi_F = 5.2$ nm and $\gamma_b = 0.21$ (down-triangles). Inset: dependence of the parameter γ_b on ξ_F.

have been kept fixed during this fitting procedure. The degeneracy related to the determination of the (ξ_F, γ_b) pairs can be removed if the superconducting critical temperature is studied for the third series of hybrids, that is $Cu_{0.38}Ni_{0.62}/[Nb/Cu_{0.38}Ni_{0.62}]_{N_{bil}}$ multilayers, with $d_S = 15$ nm and $d_F = 5$ nm, for different N_{bil} in the limit of large values of N_{bil}. The experimental points are shown in Fig. 3 by closed circles. In the same figure, by open symbols, are also shown the theoretical results obtained for four different values of the pair (ξ_F, γ_b). In order to satisfactorily reproduce the experimental data it is necessary to use for ρ_F the low temperature resistivity of a single $Cu_{0.38}Ni_{0.62}$ film 5-nm thick, $\rho_F = 135$ $\mu\Omega$ cm, resulting in $p = 0.17$. As already pointed out, in fact, the microscopic parameters describing the F-layer drastically depend on the thickness for $d_F \leq 10$ nm.[35] The values used for T_S, ξ_S, and E_{ex} are the same as above. We see that, even if a certain dispersion is present in the experimental data, they can be reproduced by the curve corresponding to the pair ($\xi_F = 6.8$ nm, $\gamma_b = 0.12 \pm 0.06$).

The same kind of analysis has been repeated for $Nb/Pd_{0.81}Ni_{0.19}$ hybrids. In this case, both the $T_c(d_F)$ and the $T_c(d_S)$ dependencies have already been successfully analyzed[36] in the frame of a different formulation of the same theoretical model.[10] For the present study a new series of samples has been deliberately fabricated. A series of $Pd_{0.81}Ni_{0.19}/[Nb/Pd_{0.81}Ni_{0.19}]_{N_{bil}}$ multilayers with N_{bil} in the range 9–12, has been in fact used to analyze the T_c versus N_{bil} dependence. The samples have $d_S = 16$ nm and $d_F = 2.2$ nm. To describe the $T_c(d_S)$ behavior for this last set of samples, we used the following parameters, namely, $\xi_S = 5.8$ nm, $p = 0.18$, and $E_{ex} = 19.8$ meV.[36] Again in the calculations we used as T_S the value obtained measuring the critical temperature of a 150-nm-thick Nb film. In this case, due to small differences in the Nb quality with respect to the one of the $Nb/Cu_{0.38}Ni_{0.62}$ series, we obtained $T_S = 8.7$ K. Keeping these values fixed we can build the degeneracy curve of pairs (ξ_F, γ_b) which reproduce with the same accuracy the $T_c(d_S)$ curve, analogously to what has been done in the case of the $Nb/Cu_{0.38}Ni_{0.62}$ system. This curve is shown in the inset of Fig. 4. Again, to remove the degeneracy the $T_c(N_{bil})$ curve for large values of N_{bil} has been studied. The experimental points are shown by closed circles in Fig. 4 where, by open symbols, the theoretical results obtained for three different values of the pair (ξ_F, γ_b) are also reported. In this case from the fit to the experimental data we obtain $\xi_F = 5.4$ nm and $\gamma_b = 0.195 \pm 0.015$. A summary of all the parameters used in this work for the two kinds of S/F multilayers is given in Table I. From the numbers obtained and reported in Table I we can estimate the specific boundary resistance for the two systems. From the expression for γ_b we have $R_B = 1.1 \pm 0.6$ fΩ m^2 for the $Nb/Cu_{0.38}Ni_{0.62}$ system and $R_B = 1.00 \pm 0.03$ fΩ m^2 for the $Nb/Pd_{0.81}Ni_{0.19}$. Both these numbers are smaller than the values obtained by other authors when measuring the specific boundary resistance of superconducting/strongly ferromagnetic systems in the CPP configuration. In fact, values around 2 fΩ m^2 for Nb/Ni (Refs. 7 and 16) and 3 fΩ m^2 for Nb/Co

TABLE I. $Nb/Cu_{0.38}Ni_{0.62}$ and $Nb/Pd_{0.81}Ni_{0.19}$ parameters used in this work to reproduce the $T_c(N_{bil})$ experimental data.

Parameter	$Nb/Cu_{0.38}Ni_{0.62}$	$Nb/Pd_{0.81}Ni_{0.19}$
T_S (K)	8.3	8.7
ξ_S (nm)	6.2	5.8
ρ_S ($\mu\Omega$ cm)	23	17
ρ_F ($\mu\Omega$ cm)	135	95
E_{ex} (meV)	11.2	19.8
ξ_F (nm)	6.8	5.4
γ_b	0.12 ± 0.06	0.195 ± 0.015
R_B (fΩ m^2)	1.1 ± 0.6	1.00 ± 0.03

(Refs. 7 and 11) have been reported. On the other hand, using the same experimental method described in this paper, we have recently obtained for a superconducting/normal metal system, namely Nb/Cu, a much smaller value for the specific boundary resistance, $R_B = 0.33$ fΩ m^2.[24] It is also interesting to compare the values estimated in this work with those reported in the literature for similar S/F systems and obtained, again, using the CPP technique. A R_B value of 10 fΩ m^2 has been recently reported for the $Nb/Cu_{0.5}Ni_{0.5}$ multilayers[17] value which is, however, unexpectedly high considering the weakly ferromagnetic nature of the alloy. Moreover, the value we have for the $Pd_{0.81}Ni_{0.19}/[Nb/Pd_{0.81}Ni_{0.19}]_{N_{bil}}$ hybrids is comparable to the boundary resistance value $R_B = 2.31 \pm 0.07$ fΩ m^2 obtained by fitting the critical current data in $Nb/Pd_{0.88}Ni_{0.12}/Nb$ Josephson junctions.[37]

Finally, it is worth noting that while for $Nb/Cu_{0.38}Ni_{0.62}$ the theoretical curves in Fig. 3 coincide in $N_{bil} = 1$, this is not true for $Nb/Pd_{0.81}Ni_{0.19}$ multilayers (see Fig. 4). This is related to the fact that the degeneracy curves (ξ_F, γ_b) for both the systems have been obtained by fitting the $T_c(d_S)$ dependence using the model of Ref. 28 which is valid in the case of $d_S > \xi_S$ and $d_F > \xi_F^*$ (here $\xi_F^* = \xi_F\sqrt{2\pi k_B T_S/E_{ex}}$). From the values reported in Table I it is evident that, while the condition $d_S/\xi_S > 1$ is fulfilled for both the systems, it is $d_F/\xi_F^* = 1.16$ for $Nb/Cu_{0.38}Ni_{0.62}$ and $d_F/\xi_F^* = 0.51$ for $Nb/Pd_{0.81}Ni_{0.19}$. This means that the conditions for having the (ξ_F, γ_b) degeneracy are strictly valid only for our $Nb/Cu_{0.38}Ni_{0.62}$ system.

III. CONCLUSIONS

In conclusion, we have studied the $T_c(d_S)$ and $T_c(d_F)$ dependence in S/F bilayers and the $T_c(N_{bil})$ behavior in $F/[S/F]_{N_{bil}}$ multilayers, using Nb as S and $Cu_{0.38}Ni_{0.62}$ and $Pd_{0.81}Ni_{0.19}$ as F. The experimental data have been interpreted in the framework of a matrix formulation of a theoretical model based on the exact solution of the Usadel equations. Due to the strong dependence of the theoretical $T_c(N_{bil})$ curves on the pairs (ξ_F, γ_b) for large value of N_{bil}, we are able to estimate the specific boundary resistance of these hybrids, without using CPP measurements which require a more complex samples preparation and, due to the extremely low values of the resistance involved, more sophisticated measurement techniques. We obtain for both systems a value of R_B of the order of 1 fΩ m^2. This number is intermediate between those measured on superconducting/normal metal and on superconducting/strongly ferromagnetic hybrids.

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