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Citation
Cirillo, C., Bell, C., Iannone, G., Prischepa, S. L., Aarts, J., & Attanasio, C. (2009). Nonmonotonic behavior of the anisotropy coefficient in superconductor-ferromagnet-superconductor trilayers. *Physical Review B*, *80*(9), 094510. doi:10.1103/PhysRevB.80.094510

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Nonmonotonic behavior of the anisotropy coefficient in superconductor-ferromagnet-superconductor trilayers

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(Received 20 May 2009; published 15 September 2009)

We have measured critical temperatures and upper critical magnetic fields as a function of the ferromagnetic layer thickness, d_F, in two different superconductor(S)/ferromagnet(F)/superconductor(S) triple layers: $Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$. We vary d_F from the 0-phase coupling to the π-phase coupling regime and find strong nonmonotonic behavior of the anisotropy coefficient $\gamma_{GL}=H_{c2\parallel}(0)/H_{c2\perp}(0)$ characterized by an initial increase, a peak, and a subsequent decrease. The peak is a manifestation of the small coupling which exists around the 0-π transition and it is qualitatively in agreement with recent theoretical predictions [B. Krunavakarn and S. Yoksan, Physica C **440**, 25 (2006)] which includes the effect of the different interface transparencies of the two systems. The experimental results demonstrate that the occurrence of the π phase strongly influences the transport properties of S/F/S systems in external fields.

DOI: 10.1103/PhysRevB.80.094510 PACS number(s): 74.45.+c, 74.78.Fk

The proximity effect in superconductor (S)/ferromagnet (F) hybrids has recently attracted a lot of interest due to the inhomogeneous nature of the superconducting order parameter in these structures.[1–3] One of the most relevant consequences of the peculiar character of the order parameter is the nonmonotonic behavior of the superconducting critical temperature T_c as a function of the thickness d_F of the F layer which has been observed in many S/F heterostructures.[4–6] Also, in the so-called S/F/S Josephson π junctions negative critical currents have been measured.[7–9] What essentially happens is that the interaction of the Cooper pairs with the exchange field E_{ex} causes the order parameter to oscillate on the F side of the interface over a distance ξ_F, the coherence length in the ferromagnet. On the other hand, on the S side, the order parameter is strongly suppressed over a distance of the order of the superconducting coherence length ξ_S, which, in conventional superconductors such as Nb, is usually of a few nanometers. In weak ferromagnetic alloys such as PdNi and CuNi, due to the smaller value of E_{ex}, ξ_F is of the order of some nanometers. In the dirty limit $\xi_F=\sqrt{\hbar D_F/E_{ex}}$ (Ref. 8) where D_F is the diffusion coefficient of the F metal. Another characteristic length introduced when studying S/F hybrids is $\xi_F^*=\sqrt{\hbar D_F/2\pi k_B T_c}$ which is a measure of the diffusive motion of the Cooper pairs in the ferromagnet and which will be needed in order to compare our data with theoretical calculations. However, the strength of the proximity effect between the S and F layers depends also on the quality of the interfaces. An important parameter in the theoretical description therefore is the interface transparency, $\mathcal{T}$.[10,11] Its influence on the behavior of the T_c both as a function of the thickness d_S of the S layer and of d_F has been studied both in Nb/CuNi and Nb/PdNi bilayers[12,13] and also, more recently, the behavior of the parallel upper critical field in these systems has been considered.[14] All these studies revealed a somehow higher value of the interface transparency in the Nb/PdNi system. Finally, not only T_c but also upper critical magnetic fields in S/F heterostructures have been theoretically predicted to oscillate as a function of d_F due to the presence of the π-phase difference between two S layers[15,16] but no experimental evidence of these predictions have been reported so far. Most of the papers devoted to upper critical magnetic fields measurements in S/F hybrids reported, in fact, on the study of coupling phenomena between the superconducting layers and on the analysis of the dimensional crossover in the temperature dependence of the parallel critical field.[17–24]

In this paper we investigate the superconducting properties of $Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers by measuring the critical temperatures and the temperature dependence of the perpendicular and parallel critical fields $H_{c2\perp}(T)$ and $H_{c2\parallel}(T)$, respectively, as a function of d_F. In particular we focused on the influence of the π-phase state on the anisotropy which is an intrinsic property of such layered structures. The behavior of $H_{c2}(T)$ and its anisotropy are in fact sensitive to the strength and the nature of the coupling between the superconducting layers.[25] The superconducting coupling between the two outer Nb layers is measured by the anisotropy coefficient $\gamma_{GL}=H_{c2\parallel}(0)/H_{c2\perp}(0)$: a stronger coupling between the superconducting layers leads to a smaller value of γ_{GL}.[26] We observe that γ_{GL} does not monotonously increase with d_F but shows a maximum in the thickness range where the π phase is formed. The comparison with the different behavior of γ_{GL} observed in $S/N/S$ trilayers (here N stands for normal metal) supports the idea that the presence of a local maximum in the anisotropy coefficient in S/F/S systems can be connected to the presence of the π phase.

Great care was paid to samples fabrication, in order to provide identical deposition conditions for all the trilayers of the series. This makes reliable the comparison between the samples of the same series, as well as the results obtained on the different trilayers systems. $Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers were deposited by dc sputtering on Si(100) substrates. The sputtering is a multi-target Ultra High Vacuum system, equipped with a load-lock chamber. The base sputtering pressure in the main chamber was in the 10^{-10} mbar range. The sputtering Argon pressure was pre-

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cisely fixed and monitored to a value of 4×10^{-3} mbar. The load-lock, with a base pressure of the order of 10^{-8} mbar, can house up to six substrates. In each fabrication run the substrates were transferred one at a time in the deposition chamber, and placed exactly in the same position to prevent the intrinsic spatial variation of the deposition rate. The latter was carefully controlled with a thickness monitor calibrated by low angle x-ray reflectivity measurements. The studied trilayers were then fabricated in groups of six, always with constant Nb thickness $d_{\mathrm{Nb}}=14$ nm and variable F thickness (1–15 nm for $\mathrm{Cu_{0.41}Ni_{0.59}}$ and 1–12 nm for $\mathrm{Pd_{0.81}Ni_{0.19}}$). In order to check the repeatability of the deposition process samples in different ranges of F layer thicknesses were deposited on purpose in the same run. This careful deposition procedure makes us confident to exclude that the results shown below are affected by samples parameters fluctuations. A very thin (1–2 nm) Al capping layer was also deposited on the top of the structures both to prevent Nb oxidation and to avoid the presence of surface superconductivity. For both the ferromagnetic alloys the Ni content which determines the magnetic strength has been checked by Rutherford backscattering analysis. The estimated Curie temperature for the $\mathrm{Cu_{0.41}Ni_{0.59}}$ alloy is $T_{Curie}\approx220$ K (Ref. 27) while $E_{\mathrm{ex}}=140$ K.[28] For $\mathrm{Pd_{0.81}Ni_{0.19}}$ we have $T_{Curie}\approx210$ K and $E_{\mathrm{ex}}=230$ K.[29] In order to compare S/F/S systems with $S/N/S$ ones, Nb/Cu/Nb trilayers with the same $d_{\mathrm{Nb}}=14$ nm have also been prepared. In this case, due to reduced pair-breaking strength of the normal metal, the Cu thickness was allowed to range up to 150 nm. Critical temperatures and critical magnetic fields were resistively measured in a ^{4}He cryostat using a standard dc four-probe technique on unstructured samples. The distance between the current pads was about 1 cm and the distance between the voltage pads was about 1 mm. T_c was taken at the 50% of the transition curves. The transition temperature of the single Nb film with $d_{\mathrm{Nb}}=28$ nm was around 8.3 K. From the slope of the perpendicular upper critical field near T_c we get for the superconducting coherence length $\xi_S\approx6$ nm. Using the expression for ξ_F reported above, the ferromagnetic coherence length in the two systems can be estimated to be $\xi_{\mathrm{CuNi}}=5.4$ nm for $\mathrm{Cu_{0.41}Ni_{0.59}}$ [with $E_{\mathrm{ex}}=140$ K and $D_F=5.3\times10^{-4}$ $\mathrm{m^2/s}$ (Ref. 28)] and $\xi_{\mathrm{PdNi}}=2.8$ nm for $\mathrm{Pd_{0.81}Ni_{0.19}}$ [with $E_{\mathrm{ex}}=230$ K and $D_F=2.3\times10^{-4}$ $\mathrm{m^2/s}$ (Ref. 29)]. With the same numbers and using $T_c=8.3$ K, we find $\xi^*_{\mathrm{CuNi}}=8.8$ nm and $\xi^*_{\mathrm{PdNi}}=5.8$ nm. Since ξ^*_F will be used to define a reduced thickness for our samples, these values mean that d_F/ξ^*_F is varied between 0 and 2 for both the CuNi and the PdNi case.

In Fig. 1 the dependence of the superconducting transition temperature on the thickness of the ferromagnetic layer is presented for both $\mathrm{Nb/Cu_{0.41}Ni_{0.59}/Nb}$ (open circles) and $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ (closed circles) trilayers. It can be seen that T_c shows a rapid drop followed by a nonmonotonic d_F dependence with a pronounced minimum at approximately 6 nm for the CuNi case or a slight minimum around 5 nm for the PdNi case. Then a saturation value of T_c is obtained at larger thickness for both the systems. It is also worth to notice that the lower T_c values measured in the $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ trilayers are probably due to both higher E_{ex} values[29] and higher interface transparency in this system with respect to $\mathrm{Nb/Cu_{0.41}Ni_{0.59}/Nb}$.[14] This peculiar $T_c(d_F)$

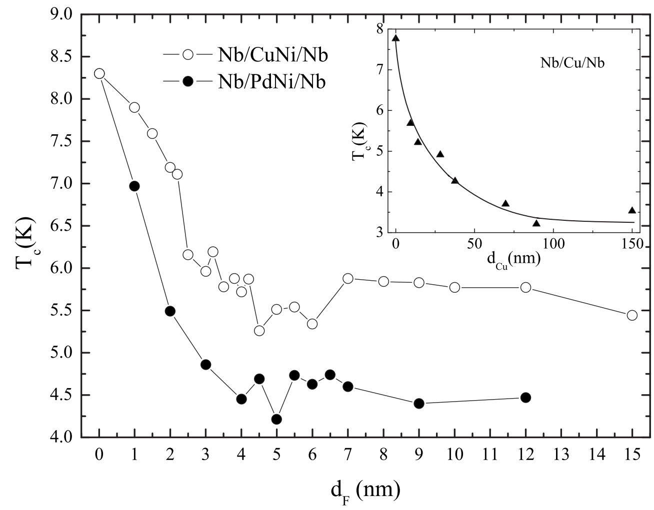

FIG. 1. Critical temperatures T_c versus the ferromagnetic layer thickness d_F for $\mathrm{Nb/Cu_{0.41}Ni_{0.59}/Nb}$ (open circles) and $\mathrm{Nb/Pd_{0.81}Ni_{0.19}/Nb}$ (closed circles) trilayers. Inset: T_c as a function of d_N in Nb/Cu/Nb system. The solid line is a guide to the eye.

behavior is a fingerprint of the 0-π phase transition in S/F hybrids,[4–7] which takes place in the thickness range where the T_c minimum occurs. As a comparison, in the inset of Fig. 1 the critical temperature dependence on the normal-metal-layer thickness, d_{Cu}, is shown for the Nb/Cu/Nb samples. Opposite to the S/F/S case, in this case a monotonous behavior of $T_c(d_{\mathrm{Cu}})$ is observed. From this result it is possible to qualitatively estimate the value of the Cu coherence length, ξ_N, the distance over which the superconductivity propagates in the N layer. Calling d^{dc}_{Cu} the distance where the two N layers are decoupled, which corresponds to the distance where T_c starts to saturate, it is possible to identify $d^{dc}_{\mathrm{Cu}}\approx2\xi_{\mathrm{Cu}}$. In our case we have $d^{dc}_{\mathrm{Cu}}\approx60$ nm so that $\xi_{\mathrm{Cu}}\approx30$ nm, which is a typical value for our Nb/Cu/Nb trilayers.[30]

In order to determine the anisotropy coefficient for all the trilayers, we measured the temperature dependence of the upper critical fields $H_{c2\perp}(T)$ and $H_{c2\parallel}(T)$. We expect the perpendicular field to be linear as a function of T, according to the expression

$$H_{c2\perp}(T)=H_{c2\perp}(0)(1-T/T_c). \tag{1}$$

On the contrary, as a consequence of the layering, decreasing the temperature $H_{c2\parallel}$ can exhibit a crossover from a linear dependence to a square-root one, namely, from a three-dimensional (3D) to two-dimensional (2D) behavior, as described by the formula

$$H_{c2\parallel}(T)=\begin{cases}H_{c2\parallel}(0)(1-T/T_c) & \text{(3D)}, \quad T>T_{\mathrm{cr}}\\ H_{c2\parallel}(0)(1-T/T_c)^{1/2} & \text{(2D)}, \quad T<T_{\mathrm{cr}}\end{cases}, \tag{2}$$

where T_{cr} is the crossover temperature. This crossover reflects the different distribution of the order parameter, which in the 3D case is spread over the entire structure while in the 2D one it nucleates in the separate thin superconducting layers. As an example, Fig. 2 shows the $H(t)$ phase diagram ($t=T/T_c$ is the reduced temperature) for the

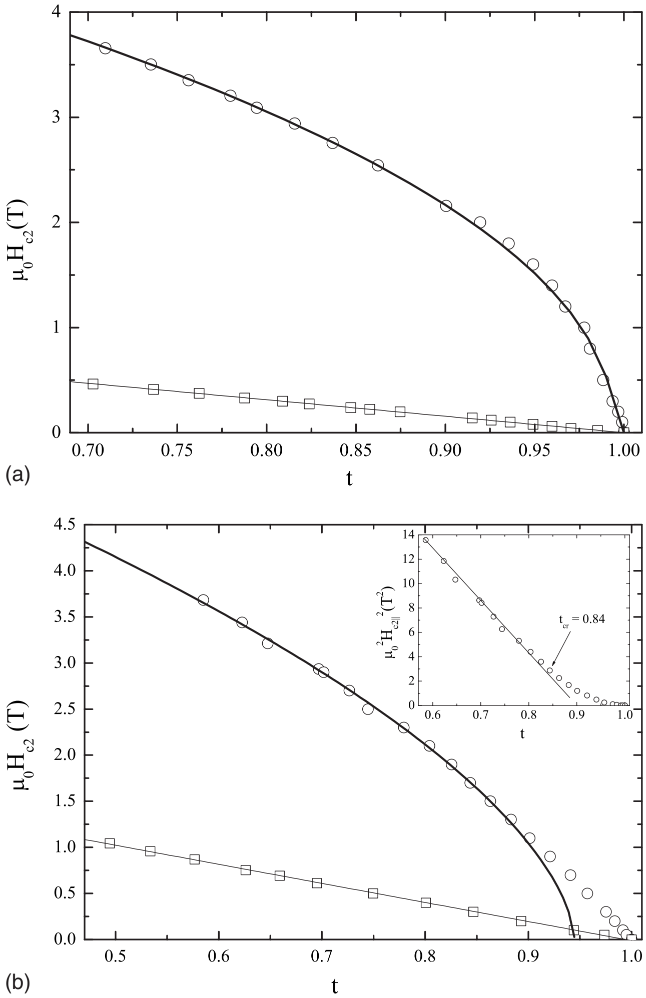

FIG. 2. (a) Phase diagrams for the Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayer with d_{PdNi}=6.5 nm. The thin (thick) solid line shows the fitted temperature dependence for $H_{c2\perp}(T)$ [$H_{c2\|}(T)$]. (b) Phase diagrams for the Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayer with d_{CuNi}=3.0 nm. The thin (thick) solid line shows the fitted temperature dependence for $H_{c2\perp}(T)$ [$H_{c2\|}(T)$]. Inset: $H^2_{c2\|}(T)$. The solid line indicates the 2D regime in this scale.

Nb/$Pd_{0.81}Ni_{0.19}$/Nb trilayer with d_{PdNi}=6.5 nm (d_F/ξ^*_{PdNi} =1.1) and for the Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayer with d_{CuNi} =3.0 nm (d_F/ξ^*_{CuNi}=0.3). For the PdNi sample [Fig. 2(a)], the perpendicular field is linear as a function of T, with $H_{c2\perp}(0)$=1.56 T, the observed temperature dependence of $H_{c2\|}$ is well described, over the entire temperature range, by the 2D expression in Eq. (2) [thick solid line in Fig. 2(a)]. The good agreement between the theoretical expression and the experimental data up to T_c indicates that the two Nb layers are completely decoupled in the whole temperature range. The value of $H_{c2\|}(0)$ obtained by fitting the experimental data using Eq. (2) is equal to $H_{c2\|}(0)$=6.60 T while the coefficient $\gamma_{GL}=H_{c2\|}(0)/H_{c2\perp}(0)$ for this sample turns out to be equal to 4.23. For the CuNi sample [Fig. 2(b)] $H_{c2\perp}(T)$ is, again, linear over the measured temperature range, with $H_{c2\perp}(0)$=2.06 T, but it is not possible for this sample to fit the $H_{c2\|}(T)$ dependence only using the 2D expression. In fact, at T_{cr}, the crossover between the 3D regime, where $H_{c2\|}(T)$ is linear and the two superconducting layers are coupled, to a 2D regime at lower temperatures, where the two Nb layers behave like two-dimensional superconducting thin films, completely decoupled by the ferromagnetic layer, occurs. If we plot $H^2_{c2\|}(T)$, as shown in the inset of Fig. 2(b), we can easily estimate the reduced crossover temperature, $t_{cr}\equiv T_{cr}/T_c$, as the point where the linear fit, which in the quadratic scale identify the 2D regime, does not match anymore with the experimental data. For the Nb/CuNi/Nb sample we then obtain T_{cr}=5.12 K. The thick solid line in Fig. 2(b) is the fit to the experimental data using the 2D expression of Eq. (2) from zero down the reduced crossover temperature t_{cr}=0.84. From this procedure we got $H_{c2\|}(0)$=7.16 T, and consequently γ_{GL}=3.48.

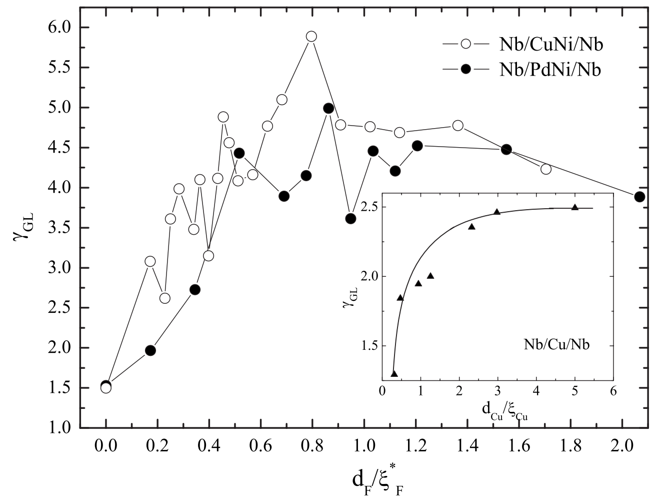

FIG. 3. Anisotropy coefficient γ_{GL} versus d_F/ξ^*_F for Nb/$Cu_{0.41}Ni_{0.59}$/Nb (open circles) and Nb/$Pd_{0.81}Ni_{0.19}$/Nb (closed circles) trilayers. Inset: γ_{GL} as a function of d_N/ξ_N in Nb/Cu/Nb system. The solid line is a guide to the eye.

In Fig. 3 $\gamma_{GL}=H_{c2\|}(0)/H_{c2\perp}(0)$ is plotted as a function of d_F/ξ^*_F for both the S/F/S trilayers, using for ξ^*_F the values calculated above. The values of the critical magnetic fields at T=0 were obtained by fitting the experimental data as described above. Again, as a comparison, in the inset of the figure the same dependence for the Nb/Cu/Nb trilayers is reported. The first thing we may notice is that the values of γ_{GL} are significantly higher for both S/F/S systems, with a clear peak around a reduced thickness of 1; this is in strong contrast with the Nb/Cu/Nb case and indicates a larger decoupling effect of the F interlayer with respect to the N case. Moreover the $\gamma_{GL}(d_F/\xi^*_F)$ dependence shows a nonmonotonic behavior for both the S/F/S systems contrary to the Nb/Cu/Nb trilayers for which the anisotropy coefficient increases monotonously showing a tendency to saturate for $d_N/\xi_N\approx 3$. We believe that the observed behavior for γ_{GL} can be related to the occurrence of the π phase, which can be assumed to set in where the T_c versus d_F curve shows a minimum.[7] In fact at the crossover from the 0 and the π phase the nature of the coupling between the two Nb layers changes, the order parameter showing a node in the center of the F layer.[31] It is then reasonable to suppose that the coupling will be strongly reduced in this regime. For this reason around the smallest coupling the anisotropy coefficient will

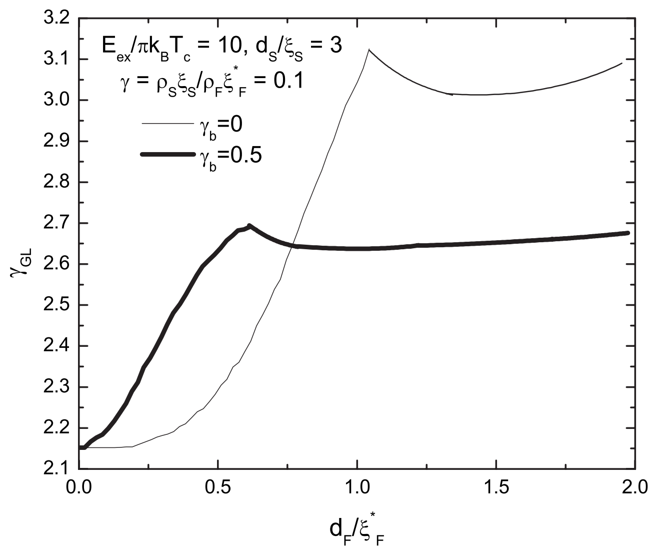

FIG. 4. Anisotropy coefficient $\gamma_{GL}=H_{c2\parallel}(0)/H_{c2\perp}(0)$ versus d_F/ξ_F^* in S/F multilayers for two different values of the parameter γ_b as deduced from data shown in Ref. 16. Both curves have been calculated for $E_{ex}/\pi k_B T_c=10$, $d_S/\xi_S=3$, $\gamma=\rho_S\xi_S/\rho_F\xi_F^*=0.1$. The lower γ_b value describes a layered S/F system with more transparent interfaces.

present a peak, which will be superimposed on the standard increase.

Indications for such nonmonotonous behavior of the anisotropy coefficient can be found in calculations of the upper critical fields in ferromagnet/superconductor layered structures (bilayers and multilayers) using the Usadel equations.[16,32] The authors calculate the reduced perpendicular and parallel critical fields at zero temperature as a function of the reduced thickness taking also into account the effect of the interface transparency $\mathcal{T}$. If the data reported in Figs. 8 and 9 of Ref. 16 are rearranged we can plot the anisotropy coefficient $\gamma_{GL}=H_{c2\parallel}(0)/H_{c2\perp}(0)$ versus d_F/ξ_F^* for S/F multilayers obtaining the results shown in Fig. 4. The two curves refer to two different values of the boundary resistivity, γ_b, but to the same values of the other parameters entering in the Usadel equations (E_{ex}, ξ_S, the superconducting layer thickness, d_S, and the resistivities of the S and F layers, ρ_S and ρ_F, respectively). γ_b is related to $\mathcal{T}$ by the relation $\gamma_b \propto \mathcal{T}^{-1}$. γ_b is infinite in the case of a completely reflecting interface ($\mathcal{T}=0$) and it is equal to 0 for a perfect transparent interface ($\mathcal{T}=\infty$). It is interesting to note that in both cases the curves are nonmonotonic showing a maximum which goes to higher d_F/ξ_F^* values for higher values of the interface transparency. Even if it should only be taken as a qualitative confirmation of our experimental data obtained on S/F/S trilayers, this result strongly supports the idea that the crossover to the π phase directly affects the coupling as measured by the critical fields. Quantitatively, however, the theoretical calculations and the experimental observations do not fully match. The behavior of γ_{GL} in the PdNi system shows a curvature, a maximum value and a thickness where the maximum is reached which are all in reasonable agreement with the calculations for full transparency. On the other hand, for the CuNi system with its lower value for E_{ex} and its lower interface transparency,[12–14] a less pronounced maximum at lower reduced thickness would be expected. Instead, the maximum value for γ_{GL} is even higher than for the PdNi case. This means that the measured decoupling is, in this case, more severe than the theory indicates. Probably, the presence of spin-flip scattering effects,[33] already invoked in the interpretation of critical current measurements in Nb/CuNi/Nb junctions,[34] should be considered in a more accurate description.

In conclusion, we have studied critical temperatures and critical magnetic fields in $Nb/Cu_{0.41}Ni_{0.59}/Nb$ and $Nb/Pd_{0.81}Ni_{0.19}/Nb$ trilayers with $d_{Nb}=14$ nm and variable d_F layer thickness. A nonmonotonous behavior of γ_{GL} has been observed as a function of d_F and it has been interpreted as due to the occurrence of the π phase in the trilayers. The different interface transparency of the two systems causes the observed shift of the maximum of γ_{GL} toward higher values of the reduced ferromagnetic thickness. The results obtained for the S/F/S systems have been compared to those obtained on $S/N/S$ trilayers where, on the contrary, γ_{GL} increases monotonously as a function of the reduced copper thickness.

This work has been partially supported (C.C. and C.A.) by the Italian MIUR-PRIN 2007 project “Proprietà di trasporto elettrico dc e ac di strutture ibride stratificate superconduttore/ferromagnete realizzate con materiali tradizionali.” It is also part (C.B.) of the research program of the “Stichting voor Fundamenteel Onderzoek der Materie (FOM),” which is financially supported by the “Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).”

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