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

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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: \( \text{Nb/Cu}_{0.41}\text{Ni}_{0.59}/\text{Nb} \) and \( \text{Nb/Pd}_{0.81}\text{Ni}_{0.19}/\text{Nb} \). We vary \( d_F \) from the 0-phase coupling to the \( \pi \)-phase coupling regime and find strong nonmonotonic behavior of the anisotropy coefficient \( \gamma_{GL} = H_{\perp\beta}(0)/H_{\perp\beta}(0) \) characterized by an initial increase, a peak, and a subsequent decrease. The peak is a manifestation of the small coupling which exists around the \( 0-\pi \) 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 \( \pi \) phase strongly influences the transport properties of \( S/F/S \) systems in external fields.

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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 \( \pi \) 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 \( \xi_F \), 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 \( \xi_S \), 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 \( E_{ex} \), \( \xi_F \) is of the order of some nanometers. In the dirty limit \( \xi_F = \sqrt{hD_F/E_{ex}} \) (Ref. 8) where \( D_F \) is the diffusion coefficient of the \( F \) metal. Another characteristic length introduced when studying \( S/F \) hybrids is \( \xi_F = \sqrt{hD_F/2\pi\kappa_B T_c} \) which is a measure of the diffuse 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, \( T \). 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 \( \text{Nb/CuNi} \) and \( \text{Nb/PdNi} \) bilayers12,13 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 \( \text{Nb/PdNi} \) 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 \( \pi \)-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 \( \text{Nb/Cu}_{0.41}\text{Ni}_{0.59}/\text{Nb} \) and \( \text{Nb/Pd}_{0.81}\text{Ni}_{0.19}/\text{Nb} \) trilayers by measuring the critical temperatures and the temperature dependence of the perpendicular and parallel critical fields \( H_{\perp\beta}(T) \) and \( H_{\perp\beta}(T) \), respectively, as a function of \( d_F \). In particular we focused on the influence of the \( \pi \)-phase state on the anisotropy which is an intrinsic property of such layered structures. The behavior of \( H_{\perp\beta}(T) \) 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 \( \gamma_{GL} = H_{\perp\beta}(0)/H_{\perp\beta}(0) \); a stronger coupling between the superconducting layers leads to a smaller value of \( \gamma_{GL} \).26 We observe that \( \gamma_{GL} \) does not monotonously increase with \( d_F \) but shows a maximum in the thickness range where the \( \pi \) phase is formed. The comparison with the different behavior of \( \gamma_{GL} \) 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 \( \pi \) 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. \( \text{Nb/Cu}_{0.41}\text{Ni}_{0.59}/\text{Nb} \) and \( \text{Nb/Pd}_{0.81}\text{Ni}_{0.19}/\text{Nb} \) 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-
cisely fixed and monitored to a value of $4 \times 10^{-3}$ 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 $d_{\text{Nb}} = 14$ nm and variable F thickness (1–15 nm for Cu$_{0.41}$Ni$_{0.59}$ and 1–12 nm for 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 Cu$_{0.41}$Ni$_{0.59}$ alloy is $T_{\text{Curie}} = 220$ K (Ref. 27) while $E_{\text{ex}} = 140$ K. For Pd$_{0.81}$Ni$_{0.19}$ we have $T_{\text{Curie}} = 210$ K and $E_{\text{ex}} = 230$ K. In order to compare S/F/S systems with S/N/S ones, Nb/Cu/Nb trilayers with the same $d_{\text{Nb}} = 14$ 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 residitively 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 $d_{\text{Nb}} = 28$ 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 $\xi_p = 6$ nm. Using the expression for $E_F$ reported above, the ferromagnetic coherence length in the two systems can be estimated to be $\xi_{\text{CuNi}} = 5.4$ nm for Cu$_{0.41}$Ni$_{0.59}$ [with $E_{\text{ex}} = 140$ K and $D_F = 5.3 \times 10^{-4}$ m$^2$/s (Ref. 28)] and $\xi_{\text{PdNi}} = 2.8$ nm for Pd$_{0.81}$Ni$_{0.19}$ [with $E_{\text{ex}} = 230$ K and $D_F = 2.3 \times 10^{-4}$ m$^2$/s (Ref. 29)]. With the same numbers and using $T_c = 8.3$ K, we find $\xi_{\text{CuNi}} = 8.8$ nm and $\xi_{\text{PdNi}} = 5.8$ nm. Since $E_F$ will be used to define a reduced coherence length for our samples, these values mean that $d_F/\xi_F$ 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 Nb/Cu$_{0.41}$Ni$_{0.59}$/Nb (open circles) and 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 Nb/Pd$_{0.81}$Ni$_{0.19}$/Nb trilayers are probably due to both higher $E_{\text{cr}}$ values and higher interface transparency in this system with respect to Nb/Cu$_{0.41}$Ni$_{0.59}$/Nb. This peculiar $T_c(d_F)$ behavior is a fingerprint of the 0–$\pi$ phase transition in S/F hybrids, 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, $d_{\text{Cu}}$, is shown for the Nb/Cu/Nb samples. Opposite to the S/F/S case, in this case a monotonous behavior of $T_c(d_{\text{Cu}})$ is observed. From this result it is possible to qualitatively estimate the value of the Cu coherence length, $\xi_{\text{Cu}}$, the distance over which the superconductivity propagates in the N layer. Calling $d_{\text{Cu}}^p$ 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 $d_{\text{Cu}}^p = 2\xi_{\text{Cu}}$. In our case we have $d_{\text{Cu}}^p = 60$ nm so that $\xi_{\text{Cu}} = 30$ nm, which is a typical value for our Nb/Cu/Nb trilayers.

In order to determine the anisotropy coefficient for all the trilayers, we measured the temperature dependence of the upper critical fields $H_{c_{2}\perp}(T)$ and $H_{c_{2}\parallel}(T)$. We expect the perpendicular field to be linear as a function of $T$, according to the expression

$$H_{c_{2}\perp}(T) = H_{c_{2}\perp}(0)(1 - T/T_c).$$

(1)

On the contrary, as a consequence of the layering, decreasing the temperature $H_{c_{2}\parallel}$ 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

$$H_{c_{2}\parallel}(T) = \begin{cases} H_{c_{2}\parallel}(0)(1 - T/T_c) & T > T_c \\ H_{c_{2}\parallel}(0)(1 - T/T_c)^{1/2} & T < T_c \end{cases}$$

(2)

where $T_c$ 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

\[ \text{FIG. 1. Critical temperatures } T_c \text{ versus the ferromagnetic layer thickness } d_F \text{ for Nb/Cu$_{0.41}$Ni$_{0.59}$/Nb (open circles) and Nb/Pd$_{0.81}$Ni$_{0.19}$/Nb (closed circles) trilayers. Inset: } T_c \text{ as a function of } d_{\text{Cu}} \text{ in Nb/Cu/Nb system. The solid line is a guide to the eye.} \]
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The value of $H_c^2(T)$ in Eq. (2) is equal to 4.23. For the CuNi sample [Fig. 2(b)] $H_{c21}(T)$ is again linear over the measured temperature range, with $H_{c21}(0)=2.06$ T, but it is not possible for this sample to fit the $H_{c2}(T)$ dependence only using the 2D expression. In fact, at $\tau_{c2}$, 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 $H_{c2}(T)$, as shown in the inset of Fig. 2(b), we can easily estimate the reduced crossover temperature, $\tau_{c2}=T_{c2}/T_c$, as the point where the linear fit, which in the quadratic scale identifies the 2D regime, does not match anymore with the experimental data. For the Nb/CuNiNb sample we then obtain $T_{c2}=5.12$ 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 $\tau_{c2}=0.84$. From this procedure we got $H_{c2}(0)=7.16$ T, and consequently $\gamma_{GL}=3.48$.

In Fig. 3 $\gamma_{GL}=H_{c2}(0)/H_{c21}(0)$ is plotted as a function of $d_F/\xi_N^2$ for both the S/F/S trilayers, using for $\xi_F^2$ 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 $\gamma_{GL}$ 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 $\gamma_{GL}(d_F/\xi_N^2)$ 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 $d_F/\xi_N^2\approx 3$. We believe that the observed behavior for $\gamma_{GL}$ can be related to the occurrence of the $\pi$ phase, which can be assumed to set in where the $T_c$ versus $d_F$ curve shows a minimum. In fact at the crossover from the 0 and the $\pi$ 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. 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 $\gamma_{GL} = H_{c2}(0)/H_{c2}(0)$ versus $d_F/\xi_F$ in S/F multilayers for two different values of the parameter $\gamma_0$ as deduced from data shown in Ref. 16. Both curves have been calculated for $E_{cs}/\pi k_B T_c = 10$, $d_S/\xi_S = 3$, $\gamma = \rho_S \xi_S/\rho_F \xi_F = 0.1$. The lower $\gamma_0$ value describes a layered S/F system with more transparent interfaces. The higher value (solid line) refers to a more opaque layered system, which is expected to have a lower interfacial transparency. The calculated curves are shown in agreement with the measured values reported in Ref. 16.

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 $d_{SF} = 14$ nm and variable $d_F$ layer thickness. A nonmonotonic behavior of $\gamma_{GL}$ has been observed as a function of $d_F$ and it has been interpreted as due to the occurrence of the $\pi$ phase in the trilayers. The different interface transparency of the two systems causes the observed shift of the maximum of $\gamma_{GL}$ 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, $\gamma_{GL}$ increases monotonically as a function of the reduced copper thickness.

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