Manipulating superconductivity through the domain structure of a ferromagnet: Experimental aspects and theoretical implications

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In the present work we study experimentally the influence that the domain structure of a ferromagnet (FM) has on the properties of a superconductor (SC) in bilayers and multilayers of $La_{0.60}Ca_{0.40}MnO_3$/Nb and FePt/Nb proximity hybrids. Specific experimental protocols that were employed in the performed magnetization measurements enabled us to directly uncover a generic property of FM/SC hybrids: In the absence of an external magnetic field, the multidomain structure of the FM promotes the nucleation of superconductivity, while its monodomain state strongly suppresses it. Our experimental findings support recent theoretical studies [A. I. Buzdin and A. S. Mel'nikov, Phys. Rev. B **67**, 020503(R) (2003); T. Champel and M. Eschrig, Phys. Rev. B **71**, 220506(R) (2005)] that suggest the formation of the so-called domain-wall superconductivity and propose that when an inhomogeneous exchange field is offered by the FM to the SC the superconducting pairs are not susceptible to pairbreaking. In contrast, when magnetic homogeneity is restored in the FM the SC's properties are strongly suppressed.

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Today, in solid-state physics the basic mechanisms that govern the fundamental structural, electronic, and magnetic physical processes of pure materials have been widely studied and in most cases well understood.[1,2] Since the potentiality of plain materials is restricted by nature, it is the exploration of artificial hybrids that in recent years has attracted great interest due to the innovative properties that they could exhibit. Such a general category of hybrids, which is the subject of the present work, refers to the combination of ferromagnets (FMs) and superconductors (SCs).[3,4]

In the field of theory it has been proposed that in FM/SC bilayers near domain walls the destruction of Cooper pairs by the exchange field is minimized so that in these regimes superconductivity may be promoted.[5] This theoretical concept was based on the fact that when superconducting pairs [which have a spatial separation of the coherence length $\xi^{SC}(T)$] are subjected to an inhomogeneous magnetization of the FM the average exchange field that they experience over $\xi^{SC}(T)$ could be strongly reduced compared to the case when the magnetization is homogeneous. Thus, the magnetic inhomogeneity of the FM effectively leads to a minimized pair-breaking effect. Since this condition is fulfilled near domain walls,[5] it is expected that in FM/SC hybrids the nucleation of superconductivity should be promoted when the FM is in a multidomain rather than in a monodomain state.[6–9] Experimentally, the multidomain state, that is inherent in all FMs above the Curie critical temperature T_c^{FM}, for $T<T_c^{FM}$ is realized near the coercive field where $m_{FM}=0$. Only few works reported on the experimental observation of this expectation. J. Aarts co-workers[10] observed sharp drops in the resistance at the coercive field of appropriately patterned $Ni_{80}Fe_{20}$/Nb bilayers. Very recently, V. V. Moshchalkov and colleagues also observed domain wall assisted superconductivity in Co–Pd/Nb/Co–Pd trilayer and $BaFe_{12}O_{19}$/Nb bilayer hybrids.[11,12]

In this work, we present magnetization data for $La_{0.60}Ca_{0.40}MnO_3$/Nb and FePt/Nb bilayers (BLs) and multilayers (MLs) hybrids. Nb has been chosen as the specific low-T_c SC since its properties are extensively studied and thus safe conclusions may be drawn from a direct comparison when the SC is in pure form and as a constituent of a hybrid. We investigated different FM materials ($La_{0.60}Ca_{0.40}MnO_3$ and FePt) and also different structures (BLs and MLs) in order to check the possible generic character of the obtained results. MLs have been investigated for one additional reason: These structures offer the opportunity for intense inhomogeneous magnetic states that may be efficiently controlled even at nanometer range by regulating the thickness of the layers that comprise the periodic structure. We stress that the case where inhomogeneous magnetization is experienced by a SC in FM/SC structures has been studied intensively in recent theoretical works.[3,8,9,13] In our work, special attention has been paid on the influence of the FM's domain state on the properties of the SC by employing minor-loops-based magnetization measurements especially designed for this purpose. *Our results clearly reveal that both in the BLs and MLs the multidomain state (inhomogeneous magnetization) of the magnetic constituent promotes superconductivity, while as a monodomain state (homogeneous magnetization) is established superconductivity is strongly suppressed.* The effect is more pronounced for the MLs because they are more magnetically inhomogeneous due to their artificially produced structural inhomogeneity. The proposed experimental methodology of minor loops offers the opportunity to study all FM/SC hybrids irrespective of their specific structure.

The $La_{0.60}Ca_{0.40}MnO_3$/Nb and FePt/Nb BLs have thickness $d_{FM}/d_{SC}=50/100$ and 20/100, respectively, while the ML/SC hybrids[14] are $[La_{0.33}Ca_{0.67}MnO_3/La_{0.60}Ca_{0.40}MnO_3]_{15}$/Nb with $[d_{AF}=4/d_{FM}=4]_{15}/d_{SC}=100$ (all values are in nm units). Details on the preparation of the laser-ablated $La_{0.60}Ca_{0.40}MnO_3$ and dc-sputtered Nb may be found elsewhere.[15,16] The FePt layers were dc sputtered on oxidized Si substrates and annealed at 600 °C for obtaining a hard magnetic phase. The MLs have $T_c^{ML}=230$ K. The Nb films have $T_c^{SC}=7$ K and 8 K for the $La_{0.60}Ca_{0.40}MnO_3/Nb$ and FePt/Nb BLs, respectively, while in the $La_{0.60}Ca_{0.40}MnO_3/Nb$ MLs they exhibit $T_c^{SC}=8.2$ K. A commercial superconducting quantum interference device (SQUID) (Quantum Design) was used for the magnetization measurements.

In all measurements the external field was applied parallel to the hybrids and the magnetization was recorded in the field-cooled (FC) procedure. In order to reveal how the domain state of a FM influences a SC, we propose a specific measuring protocol that is based on minor magnetization loops. Generally, at constant temperature $T<T_c^{SC}$, there are two extrinsic parameters that influence the behavior of the SC. First, the externally applied magnetic field H_{ex} and second, the adjacent magnetic constituent that contributes via both the stray fields that penetrate the SC and the exchange field that superconducting pairs experience as soon as they are injected from the SC into the FM. Thus, our main aim was to find an experimental way to isolate the influence of the second extrinsic parameter even in case where an external magnetic field might be present. This may be achieved in the case where the experiments are performed at *constant external magnetic field but for different magnetic states of the FM*. In order to achieve this goal, we performed successive $m(T)$ measurements when beginning from above the saturation field of the FM, we traced several minor loops by progressively increasing the field where each new minor loop started. Each one of the minor loops was accomplished at temperature $T>T_c^{SC}$. After each new minor loop, the external field was kept constant and the magnetization of the hybrid was recorded as function of temperature. All the respective $m(T)$ data belonging to a specific set of measurements *were obtained for the same value of the external magnetic field.* Representative results are shown for a ML/SC hybrid in Figs. 1(a)–1(c). In panel (a), we present the proposed measuring protocol, while in panels (b) and (c) we show data obtained for $H_{ex}=0$ Oe and $H_{ex}=-100$ Oe, respectively.

First, we will refer to the case when $H_{ex}=0$ Oe. In this set of data, we initially set $T=10\ K>T_c^{SC}$ and by starting above the saturation field of the ML we lower the applied magnetic field until a characteristic value, that we call the return field H_{ret}, is reached. Then, we start reversing the magnetic field again until the desired final value is obtained, i.e., zero in this case. At that time, we are ready to start the actual $m(T)$ measurement by lowering the temperature with typical steps of 50–100 mK until the transition of the SC is completed. When each measurement is accomplished we set again $T=10\ K>T_c^{SC}$ and we trace the next minor loop, having a higher value of the return field H_{ret}, until the external magnetic field is set back to zero. Then we are ready to perform the next $m(T)$ measurement. The whole procedure is repeated as is schematically presented in Fig. 1(a). The obtained $m(T)$ curves that are presented in panel (b) are indeed revealing. *Since these curves were obtained at* $H_{ex}=0$ Oe, *it is only the presence of the ML that affects the SC*. We clearly see that when the ML's remanent magnetization changes direction, it is also the SC's magnetization that follows (switching effect). These experiments confirm the results that have been presented very recently in Ref. 14. The switching effect survives even when a nonzero external field is applied as it is shown in panel (c). The presented data refer to the case when the external field was set to −100 Oe. Since the switching effect should occur when the ML reverses its magnetic state, we expected that this process should occur at a lower value of the return field H_{ret} for the data obtained at $H_{ex}=-100$ Oe when compared to the data obtained at $H_{ex}=0$ Oe. Indeed, this behavior is clearly observed in the data presented in Figs. 1(b) and 1(c). While for the data obtained for $H_{ex}=-100$ Oe, the SC switches its magnetization when the return field is $H_{ret}\simeq-1250$ Oe, in the data where $H_{ex}=0$ Oe the same process occurs at $H_{ret}\simeq-1700$ Oe. *These results indicate that in such systems we may use the specific state of the magnetic constituent as an efficient control parameter for the manipulation of the SC's behavior.*

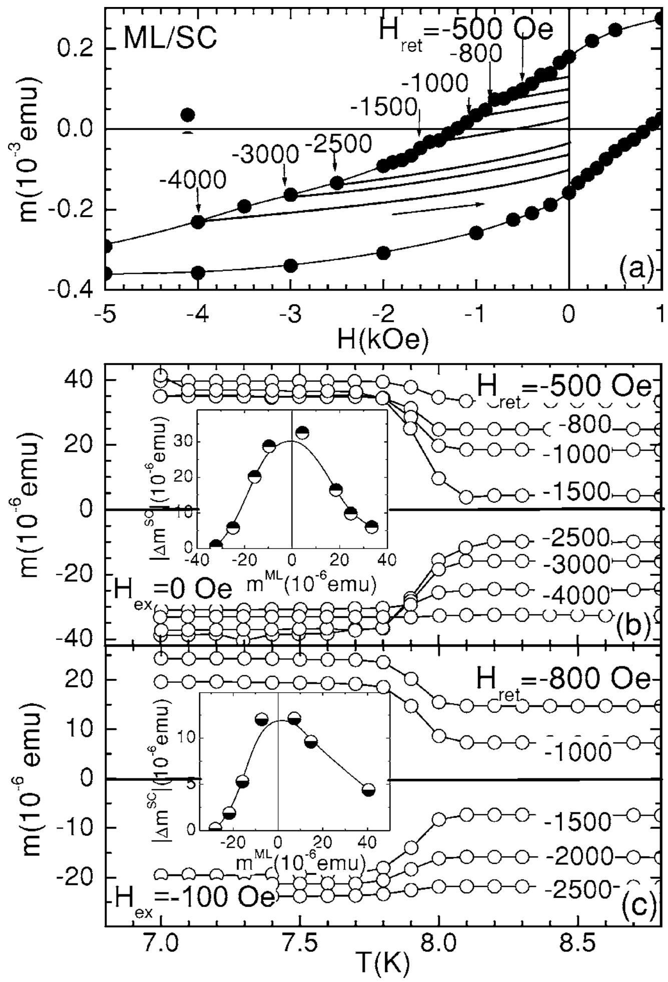

FIG. 1. (a) Schematic representation of the measuring protocol performed for a ML/SC hybrid. All minor paths end on a vertical line indicating constant external magnetic field. The respective magnetization $m(T)$ curves obtained at external fields $H_{ex}=0$ Oe and $H_{ex}=-100$ Oe are presented in panels (b) and (c), respectively. Insets present the dependence of the SC's transition height (absolute value) on the magnetization of the ML.

As already discussed in the introduction, in recent years the study of hybrid structures comprised of FMs and SCs

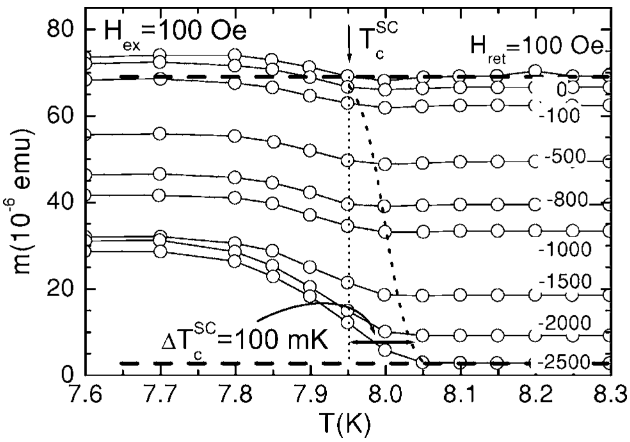

FIG. 2. Detailed $m(T)$ curves for many magnetization paths of the ML. Despite the fact that the external field is constant $H_{ex}=100$ Oe, we observe that the SC's transition temperature is lowered as the magnetization of the ML increases.

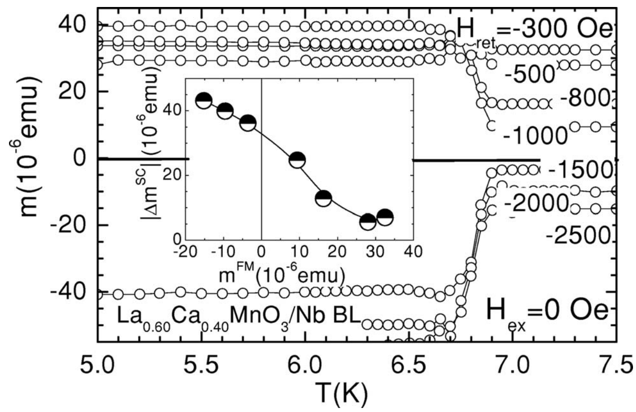

FIG. 3. Magnetization $m(T)$ curves performed according to the proposed experimental protocol for a $La_{0.60}Ca_{0.40}MnO_3$/Nb BL at external magnetic field $H_{ex}=0$ Oe. Inset presents the dependence of the SC's transition height (absolute value) on the magnetization of the $La_{0.60}Ca_{0.40}MnO_3$ layer.

have attracted the interest of experimentalists[10,11,14] not only due to the promising applications that such devices could find in the near future but also due to the vast theoretical work[5,13,17] that was done and still needs experimental feedback. An important experimental outcome of the present work that is compatible with current theoretical propositions and could offer significant information for the further experimental and theoretical examination of FM/SC hybrids is related to the influence that the domain structure of the FM has on the SC. In Fig. 1(b), we clearly see that the height of the SC's transition is enhanced significantly as the ML approaches the state of almost zero bulk magnetization where a multidomain configuration is acquired. This may be clearly observed in the insets of panels (b) and (c) where presented is the variation of the absolute value of the SC's transition height on the magnetization of the ML. Our results clearly support the recent theoretical proposals of Refs. 5–9. Until now, only one experimental report had revealed this effect by transport measurements in $Ni_{80}Fe_{20}$/Nb bilayers.[10] *Our magnetization data clearly show that the multidomain magnetic configuration that the ML acquires near-zero magnetization offers the opportunity to the superconducting pairs to sample different directions of the exchange field and thus the pair-breaking effect is only minimum.*[5–7] *As a consequence, the SC's transition height is strongly enhanced.*

Except for the transition's height, it is also the transition temperature of the SC which may be affected by the domain configuration of the FM ingredient. Figure 2 presents analytic $m(T)$ curves when many minor paths were traced for positive value of the constant external field $H_{ex}=100$ Oe. We clearly see that the maximum T_c^{SC} is observed for $m_{ML}\approx 0$. When m_{ML} increases, the critical temperature of the SC is shifted to lower values with $\Delta T_c^{SC}\approx 100$ mK. In addition, we see again that the SC's transition height is strongly reduced as a monodomain magnetic state is restored.

The same qualitative results were obtained in FM/SC BLs. Representative measurements for $La_{0.60}Ca_{0.40}MnO_3$/Nb and FePt/Nb BLs are shown in Figs. 3 and 4, respectively [the preparative minor loops as the ones presented in Fig. 1(a) for the ML/SC hybrid are not shown]. In Fig. 3, we clearly see that for external field $H_{ex}=0$ Oe, the switching effect is present and the transition's height strongly depends on the domain state of the adjacent FM layer as was observed for the ML/SC hybrids. This may be seen in its inset where presented is the dependence of the SC's transition height (absolute value) on the magnetization of the $La_{0.60}Ca_{0.40}MnO_3$ layer. The respective data obtained for a FePt/Nb BL are presented in Fig. 4 for various values of the return field, H_{ret}. Once again, we see that for $H_{ex}=0$ Oe, the transition's height of the SC strongly depends on the domain structure of the FePt layer. This is clearly presented in the inset where the SC's transition height (absolute value) is almost diminished as the FePt layer acquires a monodomain magnetic structure. Here, we should stress a strong difference between the ML/SC and FM/SC BLs that shows up when an external magnetic field is applied. In ML/SC hybrids, the domain state of the magnetic constituent still controls the SC's transition height as may be clearly seen in Fig. 1(c). In contrast, in FM/SC BLs when an external field is

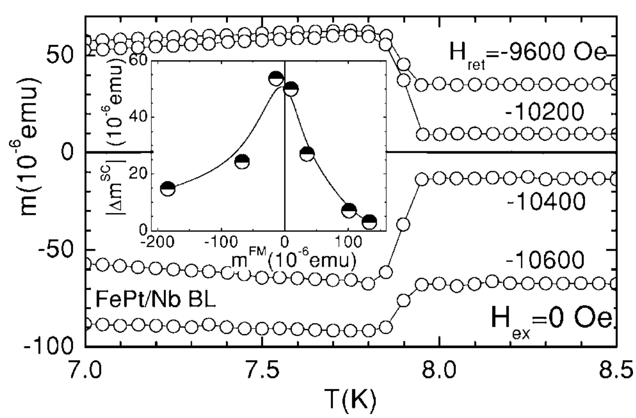

FIG. 4. Magnetization $m(T)$ curves performed according to the proposed experimental protocol for a FePt/Nb BL at external magnetic field $H_{ex}=0$ Oe. Inset presents the dependence of the SC's transition height (absolute value) on the magnetization of the FePt layer.

applied the SC's transition height is almost insensitive to the domain state of the FM layer (results not shown). We attribute this difference to the more inhomogeneous magnetic state that a ML exhibits (due to its specific structure) when compared to a single FM layer.[18]

Apart from the mechanism of the exchange interaction, we would like to discuss briefly the influence that the electromagnetic mechanism might have on superconducting pairs since the FM generates stray fields that enter the SC alongside their interface.[3,5–7] The SC's transition height is maximized near the FM's zero magnetization where the stray fields that accompany the respective magnetic domains should be randomly distributed. As a result, the average *macroscopic* stray field experienced by the adjacent SC film should be almost zero and thus could not motivate *bulk* superconductivity. However, the local contribution of the stray fields in the *mesoscopic* range cannot be neglected entirely. In this sense, since our results indicate that superconductivity is enhanced when the domain walls are maximized, we may assume that the observed effect could be equally motivated by the electromagnetic mechanism as was theoretically discussed in Refs. 6 and 7 and experimentally observed very recently in the trilayered Co–Pd/Nb/Co–Pd structures studied in Ref. 11.

Summarizing, in this work, we presented magnetization data for BL and ML hybrids consisting of $La_{0.60}Ca_{0.40}MnO_3$ and FePt combined with low-T_c SC Nb. By employing specific measuring protocols, we isolated the interplay between the domain configuration of a FM and the nucleation of superconductivity in an adjacent SC: The inhomogeneous exchange field related to a multidomain magnetic state clearly promotes the nucleation of superconductivity, while as homogeneity is restored and a monodomain magnetic state is established superconductivity is strongly suppressed. The effect is more pronounced for the MLs when compared to the BLs due to the more inhomogeneous magnetization that they exhibit intrinsically owing to their specific structure. We speculate that our experimental observations should be inherent in all FM/SC hybrids. We hope that our study will trigger further experimental and theoretical works on the possible existence of such phenomena in relative hybrid structures.

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[18] Here, we have to recall that the MLs are comprised of adjacent AF and FM layers of thickness only $d_{AF}=d_{FM}=4$ nm. In contrast, the single FM layer that is used in the BLs has thickness 50 nm and 20 nm for the $La_{0.60}Ca_{0.40}MnO_3$/Nb and FePt/Nb structures, respectively. Since the maximum size of each magnetic domain, at least in the direction along the thickness of the sample, is limited by the thickness of each respective layer, it is obvious that the MLs are more magnetically inhomogeneous when compared to the BLs.