Enhancement of Superconductivity by Exchange Bias

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In this work we study the transport properties of hybrids that consist of exchange biased ferromagnets (FMs) combined with a low-Tc superconductor (SC). Not only different FMs but also various structural topologies have been investigated: results for multilayers of La$_{1-x}$Ca$_x$MnO$_3$ combined with Nb in the form of [La$_{0.33}$Ca$_{0.67}$MnO$_3$/La$_{0.60}$Ca$_{0.40}$MnO$_3$]/15/Nb, and for more simple Ni$_{80}$Fe$_{20}$/Nb/Ni$_{80}$Fe$_{20}$ trilayers and Ni$_{80}$Fe$_{20}$/Nb bilayers are presented. The results obtained in all hybrid structures studied in this work clearly uncover that the exchange bias mechanism promotes superconductivity. Our findings assist the understanding of the contradictory results that have been reported in the recent literature regarding the transport properties of relative FM/SC/FM spin valves.

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I. INTRODUCTION

Lately, it has been shown that the combination of ferromagnets (FMs) with superconductors (SCs) offers an efficient way to bypass the natural restrictions that exist in plain materials regarding their electronic and magnetic properties. These FM/SC hybrids are promising for the design of oncoming electronics since the efficient magnetic modulation of the superconducting properties could allow the direct implementation of novel apparatus as FM/SC/FM spin valves. Although these FM/SC/FM hybrids have attracted much interest certain discrepancies that have been reported in the recent literature relent the complete understanding of the underlying physical mechanisms.

The present work offers results on the transport properties of exchange biased FMs combined with a low-Tc SC. Two quite different FMs, namely La$_{1-x}$Ca$_x$MnO$_3$ and Ni$_{80}$Fe$_{20}$, and also various structural topologies have been investigated. More specifically, MLs of La$_{1-x}$Ca$_x$MnO$_3$ combined with Nb in the form of [La$_{0.33}$Ca$_{0.67}$MnO$_3$/La$_{0.60}$Ca$_{0.40}$MnO$_3$]/15/Nb, and more simple Ni$_{80}$Fe$_{20}$/Nb/Ni$_{80}$Fe$_{20}$ trilayers (TLs) and Ni$_{80}$Fe$_{20}$/Nb bilayers (BLs) are studied. The aim of our study was to uncover the reasons responsible for the contradictory results that have been reported very recently regarding the transport properties of relative FM/SC/FM spin valves. A novel feature that is observed in the transport data obtained in all different hybrids is that the exchange bias mechanism enhances the SC’s resistive transition. This surprising experimental fact together with the magnetization data that were reported very recently, imply that the coexistence of exchange bias and superconductivity could have a positive synergetic effect.

II. PREPARATION OF SAMPLES AND EXPERIMENTAL DETAILS

The ML/SC hybrids have the form [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 (in nm units). In these samples a FM buffer layer with d$_{FM}$ = 50 nm has been used since we expected that this should act as a main reservoir for generating stray fields that, in addition to the exchange bias mechanism, could influence the SC (see inset (c) in Fig.1(a)). Information on the ML preparation may be found in Ref.11. The MLs exhibit Curie critical temperature $T_{cML} = 230$ K.

The FM/SC/FM TLs are constructed of Ni$_{80}$Fe$_{20}$/Nb/Ni$_{80}$Fe$_{20}$ (see inset (c) in Fig.3(a)) with d$_{FM}$ = 19/d$_{SC}$ = 50/d$_{FM}$ = 19 (in nm units). Also, more simple Ni$_{80}$Fe$_{20}$/Nb BLs with d$_{FM}$ = 19/d$_{SC}$ = 50 (in nm units) have been studied. For the Ni$_{80}$Fe$_{20}$ (NiFe) layers rf-sputtering was employed at 30 W in 4 mTorr Ar atmosphere (99.999% pure). We should stress that: (i) all depositions were carried out at room temperature and (ii) no magnetic field was applied during the deposition of NiFe. In order to incorporate the exchange bias only the NiFe bottom layers were treated by annealing (see10 and references therein). Afterwards, the complete TL and BL structures were deposited on top. The blocking temperature of the NiFe bottom layers is $T_B = 100$ K. Information on the preparation of the SC may be found in Ref.12. The SC critical temperature of the produced films ranges in 7.8 K <$T_{cSC} < 8.3$ K.

A commercial superconducting quantum interference device (SQUID) (Quantum Design) was used as a host cryostat. In all transport measurements the standard four-point configuration was used, the external magnetic field was applied parallel to the hybrids and the current was normal to the external field (see insets (c) in Fig.1(a) and Fig.3(a)).
FIG. 1: (a) Voltage curve $V(T)$ measured in a ML/SC hybrid from below $T_{SC}^{c} = 8.16$ K up to $T = 350$ K for warming under an external field $H_{ex} = 100$ Oe when the ML was (V) virgin (open circles) and when (BS) biased (solid circles). (b) Detailed magnetization $m(T)$ curves from $T = 2$ K up to $T = 350$ K for warming under $H_{ex} = 100$ Oe when the ML was V and BS. Inset (c) shows a schematic representation of the ML/SC hybrid structure and of the experimental configuration. Insets (d) and (e) show M(H) loops obtained at $T = 10$ K when the ML was BS and unbiased but initially saturated (UBS). The ML saturates at $H_{ML}^{sat} = 7$ kOe (inset (d)) and the loop obtained under BS conditions is shifted by 200 Oe when compared to the UBS one (inset (e)).

III. EXPERIMENTAL RESULTS

A. ML/SC hybrids

Figures 1(a) and 1(b) show in semilogarithmic plots representative transport and magnetization data that are needed for a thorough characterization of the produced ML/SC hybrids. In the lower panel we present detailed magnetization $m(T)$ curves from $T = 2$ K up to $T = 350$ K for the pure ML (prior to the deposition of the Nb layer) under $H_{ex} = 100$ Oe. The data were obtained while the ML was warmer and for two distinct cases: when the ML is initially virgin (V) and when biased (BS) [by cooling it from $T > T_{c}^{ML} = 230$ K under an external field $H_{ex} = 50$ kOe that it was lowered to $H_{ex} = 100$ Oe when the desired temperature $T = 2$ K was achieved]. First of all, we notice that the very small magnetization observed in the V curve in the low-temperature regime clearly indicates that the FM layers are coupled antiferromagnetically. The magnetization is almost constant up to the

Néel critical temperature of the AF layers $T_{c}^{AF} = 80$ K. For $T > T_{c}^{AF} = 80$ K the $V$ curve increases exhibiting a maximum at around $T = 180$ K and subsequently gets zero as $T_{c}^{ML} = 230$ K is exceeded. The BS curve is also closely related to the $V$ one presenting distinct features at the respective Néel and Curie critical temperatures $T_{c}^{AF} = 80$ K and $T_{c}^{ML} = 230$ K. As expected, above $T_{c}^{ML} = 230$ K the magnetization gets zero. For $T_{c}^{AF} = 80$ K < $T < T_{c}^{ML} = 230$ K the BS curve attains a constant value, while for $T < T_{c}^{AF} = 80$ K it exhibits high values. The respective insets (d) and (e) show M(H) data for the ML obtained at $T = 10$ K. Inset (d) shows a representative loop in an extended field regime, while inset (e) focuses in the low-field regime for the M(H) loops obtained when the ML was biased (BS) and unbiased but initially saturated (UBS). These data clearly show that the exchange bias mechanism controls the magnetic behavior of the ML. The characteristic features observed in the $m(T)$ curves of the lower panel are also visible in the respective $V(T)$ ones presented in the upper panel. Its inset (c) shows a schematic representation of the ML/SC hybrid structure and of the experimental configuration. As may be seen the resistive transition of the SC occurs at $T_{SC}^{c} = 8.16$ K.

The detailed magnetization data that were presented in Refs. 11,12 for the same ML/SC hybrids revealed that the exchange bias mechanism influences the SC’s magnetic behaviour. In order to investigate if, except for the magnetic behavior, the exchange bias could influence the SC’s transport properties we performed resistivity measurements. Representative data are shown
in Figs.2(a) and 2(b) revealing that the exchange bias mechanism clearly affects the current-carrying capability of our ML/SC hybrids. Measurements were performed while lowering either the applied magnetic field (upper panel) or the temperature (lower panel) for two different conditions: when the ML was initially BS (dotted curves) and when UBS (solid curves) [see also inset (e) in Fig.1(b)]. We observe that the BS curves are placed below the UBS ones. This is a clear evidence that superconductivity is enhanced when the SC experiences the exchange bias imposed by the adjacent ML.

B. FM/SC/FM TL and FM/SC BL hybrids

In order to investigate the generic character of this result we examined another quite different FM constituent, namely NiFe combined in two NiFe/Nb/NiFe TL and NiFe/Nb BL topologies with the SC. Figures 3(a) and 3(b) show in semilogarithmic plots representative results that reveal the existence of exchange bias in a NiFe/Nb/NiFe TL. We remind that only the bottom NiFe layer exhibits exchange bias so that it is called BFM ("B" stands for biased), while the as-deposited top NiFe layer is noted as PFM ("P" stands for plain). Panel (a) presents the resistive curve from below $T_c^{SC} = 7.81$ K up to $T=120$ K, while in panel (b) presented are the zero field cooled and field cooled magnetization $m(T)$ curves under $H_{ex} = 10$ Oe when the sample is virgin. Inset (d) of panel (b) presents $m(H)$ loops obtained at $T=8$ K when the TL was virgin and BS [biasing is achieved by cooling the TL from $T>T_B = 100$ K down to $T=8$ K under an external field $H_{ex} = 500$ Oe].

Measurements as a function of temperature for different magnetic states of the TL are shown in Figs.4(a)-4(c). In nice agreement with the ones presented in Fig.2 for the ML/SC hybrid, these data reveal that the exchange bias clearly promotes the resistive critical temperature of the TL as for instance may be seen in panel (a). In panel (b) although both curves were obtained for BS TL they refer to different applied fields, $H_{ex} = 50$ and $50$ Oe. Thus, these measurements refer to different magnetization configuration of the outer NiFe layers: at $H_{ex} = 50$ Oe the layers are antiparallel and at $H_{ex} = 50$ Oe they are parallel (see the BS loop in inset (d) of Fig.3(b)). This information will help us to discuss the recent discrepancies that have been reported in relevant FM/SC/FM spin valves. Since in all measurements presented in Figs.2(b) and 3(a)-3(b) the observed temperature shift is very small (but, at least, comparable to the ones presented in Refs.5,6,7,8,9) we performed some test measurements in order to ensure that these shifts are motivated by the physics of the studied systems and are not coincidental. The results of panel (c)
serve this aim since they clearly show that at the symmetric points $H_{ex} = -300$ and 300 Oe of the virgin TL the obtained V(T) curves clearly coincide as they should (see the virgin loop in inset (d) of Fig. 3(b)).

Figure 5(a) shows V(H) curves for a second TL obtained close to its $T^SC = 7.89$ K. Both virgin and BS curves are shown. Firstly, the virgin curves present a broad peak around zero field. As we see from the virgin m(H) curve in inset (d) of Fig. 3(b) at this field region the TL’s magnetization gradually reverses. This result is identical to the one presented by V. Peña et al. in Ref. 8. More importantly, here we clearly demonstrate that this peak may be almost entirely suppressed by the application of the exchange bias. Figure 5(b) shows the respective V(H) curves for a BL obtained close to its $T^SC = 8.26$ K. In both the TL (panel (a)) and the BL (panel (b)) we see that while in the normal state the virgin and BS curves coincide, as we enter in the superconducting state these curves diverge, with the BS curve placed significantly below the virgin one.

**IV. DISCUSSION**

Let us now discuss the contradictory experimental results that have been reported in the recent literature for relevant spin valves. The concept of a SC spin valve is based on a FM/SC/FM TL structure where the nucleation of superconductivity can be controlled at will by the relative magnetization configuration of the outer FM layers. J.Y. Gu et al. were the first who reported on the experimental realization of a Ni$_2$Fe$_{18}$Cu$_{0.47}$Ni$_{0.53}$/Nb/Cu$_{0.47}$Ni$_{0.53}$/Ni$_2$Fe$_{18}$ spin valve. In that work the exchange bias mechanism was used since an additional Fe$_{50}$Mn$_{50}$ layer was introduced in order to control the magnetization of the one FM layer. It was observed that when the magnetizations of the two FM layers were antiparallel (parallel) the resistive transition of the SC was placed at higher (lower) temperatures. Soon after, A. Potenza and C.H. Marrows, and also I.C. Moraru et al. by employing also the exchange bias mechanism confirmed the results of Ref. 8. Contrary to those results, in the experiments of V. Peña et al and of A. Yu. Rusanov et al. where La$_{0.7}$Ca$_{0.3}$MnO$_3$YBa$_2$Cu$_3$O$_7$/La$_{0.7}$Ca$_{0.3}$MnO$_3$ and Ni$_{50}$Fe$_{20}$/Ni$_{50}$Fe$_{20}$ TLs were studied respectively, the opposite behavior was observed. Both works reported that the antiparallel (parallel) magnetization configuration of the FM layers suppresses (enhances) superconductivity. We stress that in Refs. 8,9 the exchange bias mechanism was not employed. Accordingly to our results when the SC experiences the exchange bias its resistive transition is enhanced. This finding could explain the discrepancies reported in Refs. 8,9. The results presented in Fig. 3(b) reveal that the antiparallel magnetization configuration of the outer NiFe layers suppresses superconductivity in agreement with Refs. 8,9.

Finally, exchange bias is related to unidirectional magnetic anisotropy and phenomenologically it can be viewed as a parameter that controls the nucleation and orientation of magnetic domains in a FM; a key parameter for the FM/SC/FM spin valves. Since we observed that the parallel magnetic configuration enhances superconductivity we may assume that exchange bias also promotes superconductivity by promoting the alignment of the magnetic domains over the whole FM. Ultimately, this could somehow be related to the existence of a spin-triplet supercurrent since, according to basic knowledge under such conditions a spin-singlet one should be seriously suppressed due to the detrimental influence of the FM’s exchange field. More specific experiments are needed to confirm this hypothesis.

**V. CONCLUSIONS**

Summarizing, we presented transport data in [La$_{0.33}$Ca$_{0.67}$MnO$_3$/La$_{0.60}$Ca$_{0.40}$MnO$_3$]$_{15}$/Nb, Ni$_{50}$Fe$_{20}$/Ni$_{50}$Fe$_{20}$ and Ni$_{50}$Fe$_{20}$/Nb hybrids. A novel finding that we observed is that in all different structures the exchange bias mechanism improves the resistive transition of the FM/SC hybrid. Since this feature is independent of the specific FM material and of the structural topology we believe that it should be generic in all FM/SC hybrids. Our experiments assist the understanding of the discrepancies that have been reported in the recent literature regarding relative FM/SC/FM spin valves.
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