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Magnetic field dependence of the proximity-induced triplet superconductivity at ferromagnet/superconductor interfaces

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Long-ranged superconductor proximity effects recently found in superconductor-ferromagnetic (S-F) systems are generally attributed to the formation of triplet-pairing correlations due to various forms of magnetic inhomogeneities at the S-F interface. In order to investigate this conjecture within a single F layer coupled to a superconductor, we performed scanning tunneling spectroscopy on bilayers of $La_{2/3}Ca_{1/3}MnO_3$ (LCMO) ferromagnetic thin films grown on high-temperature superconducting films of $YBa_2Cu_3O_{7-\delta}$ or $Pr_{1.85}Ce_{0.15}CuO_4$ under various magnetic fields. We find a strong correlation between the magnitude of superconductor-related spectral features measured on the LCMO layer and the degree of magnetic inhomogeneity controlled by the external magnetic field. This corroborates theoretical predictions regarding the role played by magnetic inhomogeneities in inducing triplet pairing at S-F interfaces.

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Proximity effects in superconductor-ferromagnetic (S-F) hybrids became the subject of intensive research in recent years, partly due to observations of long-range spin-polarized supercurrents in superconductor-ferromagnetic-superconductor (S-F-S) Josephson junctions, signifying the appearance of a proximity-induced spin-triplet-pairing order in the F layer. It is well known that ferromagnetism and spin-singlet superconductivity are two inimical orders as one is associated with electrons of parallel spin alignment and the other with Cooper pairs formed from electrons with antiparallel aligned spins. Consequently, the proximity effect (PE) in S-F junctions is expected to be short ranged due to the exchange field (E_{ex}) in the F that acts to dephase the two electrons with opposite spin sign [1,2]. This leads to a very short penetration depth of superconducting order into the F, on a length scale of $\xi_F = \sqrt{\hbar D/2E_{ex}}$, where D is the diffusivity in the F. For strong ferromagnets, $\xi_F \sim 1$ nm, much shorter than the typical penetration depth of the S order into a normal metal $\xi_N = \sqrt{\hbar D/k_B T}$, that can be as large as 100 nm at low temperatures.

Despite the expected short coherence length of Cooper pairs in F materials, over the past decade, experiments on S-F-S Josephson junctions with the half-metallic ferromagnet (HMF) (CrO_2) [3–5], intermetallic (Cu_2MnAl) [6], and metallic (Co) [7,8] barriers have revealed evidence of supercurrents in F barriers much thicker than ξ_F. Equal-spin Andreev reflections and long-range coherent transport were also found in $YBa_2Cu_3O_{7-\delta}/La_{2/3}Ca_{1/3}MnO_3$ (YBCO/LCMO) multilayers through the observation of McMillan-Rowell resonances [9], and signatures of a triplet in $La_{0.7}Sr_{0.3}MnO_3$/YBCO/$La_{0.7}Sr_{0.3}MnO_3$ trilayers appeared in the magnetic field evolution of the critical temperature and conductance spectra [10]. In addition, scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS) experiments [11,12] provided evidence of long-ranged PE in HMF LCMO films for which ξ_F is estimated to be smaller than 1 nm ($E_{ex} \sim 3$ eV and the Fermi velocity is rather small $\sim 7 \times 10^7$ cm/s) [13]. Superconducting-related features were observed in the tunneling spectra measured on LCMO layers as thick as ∼30 nm, much larger than ξ_F, in bilayers of LCMO/(100)YBCO and LCMO/$Pr_{1.85}Ce_{0.15}CuO_4$ (PCCO) [11,12]. However, such effects were not found for LCMO/(001)YBCO bilayers [14] as discussed in the Supplemental Material [15].

The mechanisms proposed for the long-ranged PE involve the conversion of singlet state Cooper pairs into an equal-spin-triplet state in the F, which is insensitive to the ferromagnetic exchange field. It has been suggested that such a conversion process can occur due to some form of magnetic inhomogeneity, such as a domain wall [16] or a spin-active F-S interface [17,18]. Bergeret *et al.* [19] showed theoretically that rotation of the magnetization inside a domain wall, for example, can promote the formation of an equal-spin-triplet state. Similarly, in a mechanism proposed by Eschrig and Löfwander [17], an F-S interface can give rise to a triplet state with zero projection in spin space on the magnetization axis of the interface. If the magnetizations at the F-S interface and inside the F layer are noncollinear, the triplet state at the interface has a nonzero spin projection on the F-layer interior magnetization, and the corresponding equal-spin-triplet component decays in the F on a length scale of ξ_N. For an excellent review on this issue, see Ref. [20]. Furthermore, these mechanisms predict that the induced superconductor order parameter (OP) can have an orbital symmetry which is even (s or d wave) or odd (p or f wave), maintaining fermionic antisymmetry

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by a suitable odd or even dependence on the Matsubara frequency. Because anisotropic OPs (e.g., d wave or p wave) are sensitive even to nonmagnetic disorder, it is expected that the superconducting correlations should predominately be carried into the F by an odd frequency s-wave component, whereas, lower symmetry OPs should appear less abundantly. Our previous STM measurements on LCMO/YBCO [11] and LCMO/PCCO [12] bilayers are consistent with this prediction; the tunneling spectra revealed mainly proximity gaps over large regions on the LCMO surface and, to a much lesser extent, zero-bias conductance peaks (ZBCPs), which are known to be associated with anisotropic sign-changing OPs, such as p and d wave.

Despite ample evidence for PE-induced triplet pairing at S-F interfaces and some recent experiments which clarify the role of magnetic inhomogeneity in engineered magnetic multilayers [7,8,21], it is still not well established experimentally whether the intrinsic magnetic inhomogeneity within a *single* ferromagnetic material proximity coupled to a superconductor plays an important role in generating equal-spin-triplet components as suggested by theory [17,18]. Here, we report STM and STS measurements in a magnetic field showing that the degree of magnetic homogeneity within a single F layer can greatly affect the magnitude of the superconducting-related spectral features in LCMO proximity coupled to YBCO or PCCO. Our findings show that magnetization misalignment between different regions around S-F interfaces is key to generating a long-range PE.

STM and STS measurements were performed at 4.2 K on two series of epitaxial bilayers grown by pulsed laser deposition, consisting of LCMO films with thicknesses between 10 and 20 nm ($\gg \xi_F$) deposited on two different high-temperature superconductor films. The first are optimally (hole) doped a-axis (100)YBCO films, 135-nm thick, grown on (100)SrTiO$_3$ substrates, and the second are 300-nm-thick (electron-doped) PCCO films, deposited on (001) NdGaO$_3$ substrates. In both cases, the S and LCMO films were grown consecutively without breaking vacuum, showing a magnetic transition at ~250 K and superconducting transitions at 88 K (YBCO) and 17 K (PCCO). For further details, see the Supplemental Material [15]. We also acquired tunneling spectra on control samples of bare 15- and 20-nm-thick LCMO grown directly on substrates of (100)LaAlO$_3$. The corresponding results are described in the Supplemental Material [15].

The spatial distribution of the superconducting-related spectral features found in our previous studies [11,12] in zero magnetic field was reproduced in the samples studied here. Namely, superconducting-related spectral features (mainly gaps) were observed over large regions, hundreds of nanometers in size (much larger than the width of domain walls in LCMO, ~20 nm) [22]. Figure 1 presents tunneling dI/dV versus V spectra featuring gaps in the quasiparticle density of states (DoS) on a 17-nm LCMO/YBCO bilayer as a function of applied magnetic field. The spectra were acquired without changing the STM (current and voltage) settings except for the magnetic field, which was applied perpendicular to the S-F interface. Before applying a magnetic field, the spectrum is gapped with a normalized ZBC of 0.73. When applying a magnetic field of 75 mT, the gap becomes more pronounced with the normalized ZBC decreasing to 0.64, indicating a reduction in the quasiparticle DoS at the Fermi energy. It is important to note that when applying a magnetic field perpendicular to the easy axis, which lies in plane in our sample, the saturation field of the LCMO films is found to be ~300 mT [23]. By further increasing the magnetic field, the ZBC rises until it reaches 0.85 at 360 mT. The inset of Fig. 1 shows the normalized ZBC taken from each curve as a function of the magnetic field.

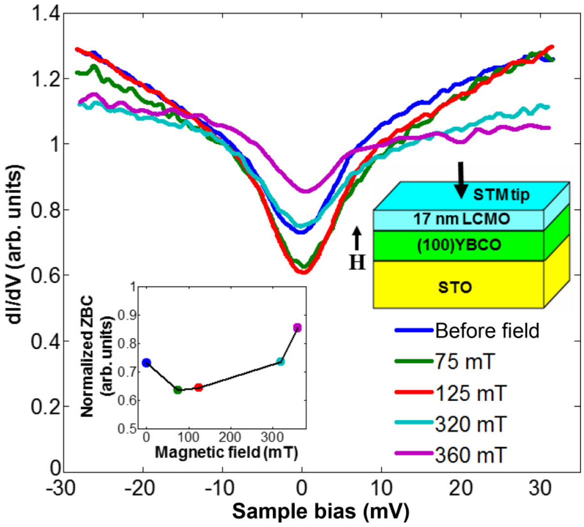

FIG. 1. (Color online) Normalized (to the gap edge) tunneling dI/dV vs. V spectra at 4.2 K acquired on a 17-nm LCMO/(100)YBCO sample in different magnetic fields applied perpendicular to the interface. Left inset: The normalized zero-bias conductance of the superconductor proximity gap as a function of the magnetic field. Note the nonmonotonic dependence of the ZBC on the magnetic field. Right inset: Scheme of the bilayer film and measurement configuration. The error bars are smaller than the dot size.

A similar nonmonotonic dependence of the proximity-induced gap on the magnetic field was found for the 14-nm LCMO/YBCO bilayer as shown in Fig. 2. The inset of Fig. 2 shows that the magnitude of the gap hardly changes when the applied field is reduced from 180 mT to zero and then increased to 150 mT. This behavior implies that the magnetization distribution remained intact (at least in the measurement region) during the field recycling process, which took place over a time scale of ~1 min. The effect of such magnetic viscosity is discussed in the Supplemental Material [15].

Another example of the evolution of superconducting-related spectral features in a magnetic field is shown in Fig. 3. Here, each curve is an average over tens of similar stable spectra all acquired at the same place on LCMO. At zero field, we observed a peak slightly shifted toward negative bias and superimposed over an asymmetrical conductance background. In most cases, eliminating the background conductance by subtracting a polynomial fit to the curve excluding the central peak reduces the peak shift from about 2 mV to nearly zero bias as shown, for example, for the spectrum acquired at 490 mT [inset of Fig. 3(a)]. We, therefore, refer to such peaks as ZBCPs, although in some spectra, a small shift of up to 1 mV remains even after background subtraction, possibly due to asymmetric splitting of the ZBCP (see the Supplemental Material [15]), which is too small to be resolved.

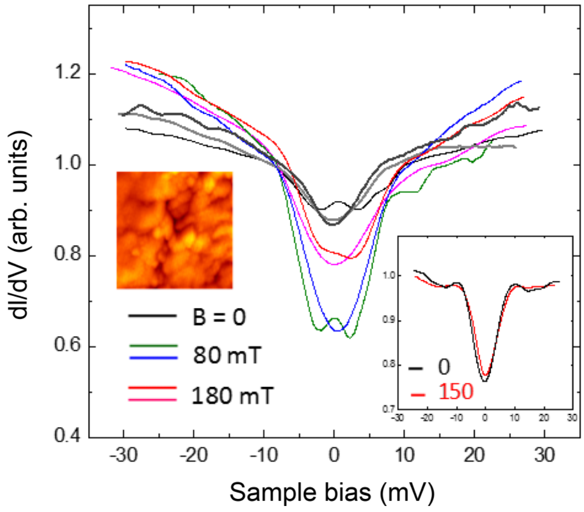

FIG. 2. (Color online) Normalized (to the gap edge) tunneling spectra at 4.2 K in different magnetic fields as indicated on a 14-nm LCMO/(100)YBCO bilayer, typical of those measured over a $50 \times 50\ \text{nm}^2$ region shown by the topographic image (left inset). All spectra exhibit proximity gaps and, in some cases, a signature of a ZBCP. The right inset shows two spectra, one measured after reducing the field to zero (from 180 mT) and the other after subsequently ramping the field from zero to 150 mT.

As the field was increased to 80 mT, the ZBCP became more pronounced, and the curve hardly varied as the field was increased further to 120 mT. However, after further increasing the field to 175 mT, the ZBCP diminished in height and became less pronounced than at zero field. This behavior is reminiscent of that of the gap shown in Fig. 1. When the field was lowered back to 120 mT, the ZBCP increased again (not shown). A nonmonotonic behavior was also observed in the course of the measurement at higher fields as can be seen in Fig. 3(b). Namely, the small ZBCP at 225 mT increased gradually up to 410 mT but eventually faded away entirely at ~585 mT. After lowering the field to zero again, the spectra remained unchanged from those acquired at 585 mT. The ZBCP height extracted from each curve as a function of the magnetic field is shown in the inset of Fig. 3(b).

An effect of the applied field on superconducting-related features was observed also for a 15-nm LCMO/PCCO bilayer as shown in Fig. 4. Here, we present spectra acquired after zero-field cooling (blue and green curves) and after field cycling to 180 mT and back to zero field (reddish curves). All spectra were measured over the same region using identical STM settings. Figure 4(a) shows that the gap feature is about twice as deep after applying the field as compared to the zero-field gap, and Fig. 4(b) shows that the ZBCP has almost doubled also after application of the field. We attribute the difference between the spectra acquired before any field was applied and after the field was set back to zero (Fig. 4) to magnetization hysteretic effects that are discussed in the Supplemental Material [15]. There, we show evidence for dynamic (time-dependent) superconductor PE features in the spectra acquired on LCMO in response to a magnetic field (Fig. S1). The temporal evolution of these features, some of which were not abundantly found, resembles that of magnetization-related phenomena observed in thin LCMO films, both in our STM measurements and in a previous [24] magnetotransport study. This behavior provides further evidence to support our claim that magnetic inhomogeneity plays a key role in generating triplet pairs in LCMO.

The mechanisms suggested for the induction of a triplet superconductor OP inside a ferromagnet require some form of magnetic inhomogeneity. The corresponding noncollinearity in the magnetization of nearby regions in the F has been

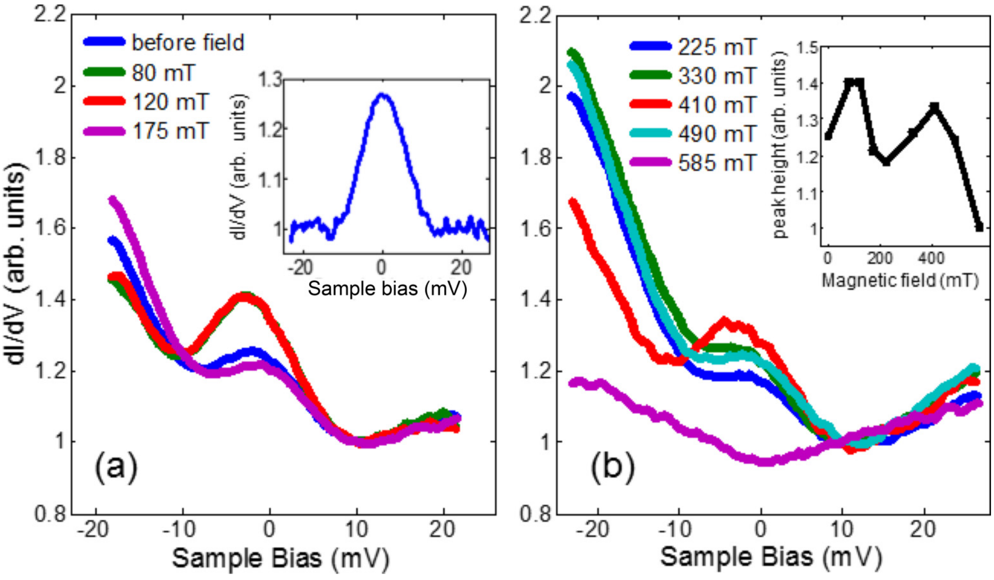

FIG. 3. (Color online) Normalized (to the right dip) tunneling dI/dV vs. V spectra at 4.2 K acquired on a 17-nm LCMO/(100)YBCO sample showing the evolution of the ZBCP with magnetic fields in the regimes of (a) 0–175 mT and (b) 225–585 mT. By subtracting the asymmetric background conductance as explained in the main text, the bias shift of the ZBCP decreases practically to zero as seen in the inset of (a) for a spectrum acquired at 490 mT. The magnitude of the ZBCPs shows a complex dependence on the magnetic field as portrayed by the inset of (b) where the peak height is plotted as a function of the magnetic field; the black line is a guide to the eye.

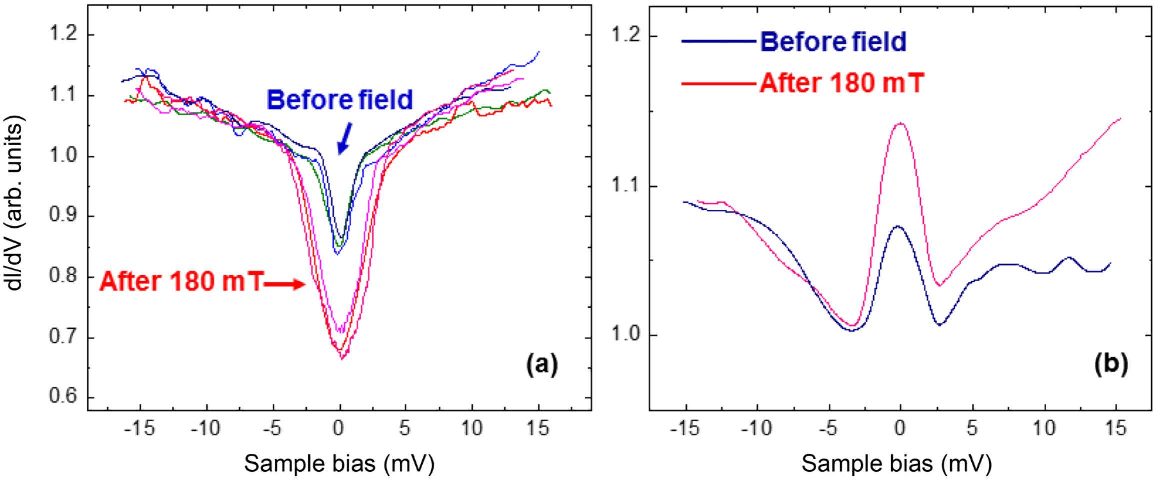

FIG. 4. (Color online) Normalized tunneling dI/dV vs. V spectra at 4.2 K acquired from a 15-nm LCMO/(001)PCCO bilayer at zero field. The blue and green curves were measured right after zero-field cooling, whereas, the reddish curves were measured right after ramping the field to 180 mT and back to zero. It is evident that the superconductor-related features, either (a) the gap or (b) the ZBCP, are enhanced due to the field cycling.

predicted theoretically to play an important role in the creation of a triplet-pairing component in F-S structures [25,26]. The magnetic inhomogeneity may be associated, e.g., with domain walls [16] or with roughness or strain at the interface that may cause misalignment of the interface magnetization with that of the F-layer interior [17,18]. Another form of magnetic inhomogeneity specific to (001)YBCO/LCMO interfaces was reported by Chakhalian *et al.* [27] where a layer with suppressed magnetization on the LCMO side extends 1 nm from the interface and is antiferromagnetically coupled to a 2-nm-thick spin-polarized layer in the YBCO. Since the magnetizations of both layers are antiparallel, a small magnetic field will cause them to misalign. A similar characterization of (100)YBCO/LCMO or PCCO/LCMO bilayers, such as the one we studied, is still lacking.

As we have shown in previous papers [11,12] and confirm in the present Rapid Communication, superconductor PE-related features were found in the tunneling spectra (in most cases) on very large areas compared to domain walls of LCMO. We thus assume that some form of widespread magnetic inhomogeneity involving misalignment of magnetization in adjacent regions is at play in our two systems. Applying a magnetic field to samples with such misalignment will eventually tend to align the magnetizations in both regions along the direction of the applied field. This, however, does not imply that magnetizations in such adjacent regions will tend to align in the same direction at low fields. Specific to our samples, it is important to note that the magnetization easy axis of LCMO lies in plane [23], whereas, the magnetic field was applied perpendicular to the plane. Also, the anisotropy constants and magnetic viscosity in different regions are highly likely to differ. Thus, within a finite time, a low field will tend to rotate the magnetization axis out of the plane in adjacent regions but to different extents, depending on the magnetic properties of each region. Since it is likely that the initial magnetizations are predominantly in plane, a low perpendicular field will increase, in many configurations, the misalignment between the adjacent regions. However, at high enough fields, the magnetizations in both regions are expected to align along the applied field, thus, reducing the net inhomogeneity. According to this scenario, the application of a magnetic field is expected to result in an initial enhancement of the triplet-pairing state in the F at low fields, followed by its suppression at higher fields. We note that a maximum in the triplet component penetration is anticipated when the magnetizations in the two regions become perpendicular to each other. Since we do not know the exact magnetization configuration throughout the measurement, we cannot perform a quantitative analysis of the PE as a function of misalignment or size of the magnetic regions. However, as shown by Figs. 1–3, it is clear that the proximity-induced gaplike features and ZBCPs are affected by the magnetic field in a manner consistent with theory and the field range in which the changes take place are in accord with the magnetization curves presented in the Supplemental Material [15]. Figure 3 shows a more complex behavior where, at even higher fields, an additional enhancement and subsequent reduction in the ZBCP are observed. This behavior can possibly be explained by considering more complex initial magnetization structures. These include, for instance, two regions with magnetization directions making an obtuse angle with the magnetic field or three regions with different magnetic susceptibilities. In any case, at a high field (585 mT), where the magnetization is expected to become homogeneous, the ZBCP also disappears, and the spectra do not manifest any superconducting-related features. Figure 4 focuses on the enhancement at low fields, which persist for some time due to magnetization viscosity, even after the field is reduced back to zero.

Other scenarios can account for the nonmonotonic dependence of the superconductor-related features on the magnetic field. For instance, domain-wall motion due to the applied field may locally change the magnetic structure and, thus, the magnitude of the triplet-pairing PE in the vicinity of the STM tip. However, the observation of superconducting OP penetration on a much larger scale [11,12], possibly due to interface versus F-interior magnetization misalignment, may mask this effect, which is more local in nature. Adjacent regions, either magnetic microdomains or crystallites, possibly separated by antiphase boundaries, may constitute another form of magnetic inhomogeneity [24,28]. These regions are coupled to one another via dipole-dipole interaction and are, thus, polarized antiparallel to each other at zero field. In response to the application of a magnetic field, the magnetization orientations in these regions will evolve from being in plane and antiparallel to become aligned parallel to the applied field. Thus, the magnetization noncollinearity will initially increase and, subsequently, will decrease with a corresponding enhancement and suppression of the triplet state.

The changes in ZBCPs due to magnetic field application in our LCMO/YBCO bilayers may partially be associated with corresponding effects observed on bare (110)YBCO [29] and $Y_{0.95}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ [30]. Here, changes in ZBCP height and splitting thereof in response to a magnetic field were attributed to a complex order parameter. The effects observed in these experiments occurred at fields of ~1 T, whereas, we observed considerable effects already at 70 mT. However, if one takes into account local fields inside the LCMO or close to it, such intensities are attainable. Consequently, if a complex OP emerges in either the YBCO or the LCMO, it may also contribute to changes we observe in response to the magnetic field, including splitting of ZBCPs (see the Supplemental Material [15]).

To summarize, we have studied the magnetic field dependence of an induced triplet-pairing order parameter in ferromagnetic LCMO films, much thicker than ξ_F, coupled to either YBCO or PCCO using scanning tunneling spectroscopy. The magnitude of the superconducting-related spectral features, proximity gaps and ZBCPs, showed a nonmonotonic dependence on applied magnetic field, following the anticipated concomitant evolution of magnetic inhomogeneity in the LCMO. Our data, thus, indicate that the local magnetization configuration largely controls the formation of an induced triplet order parameter in the LCMO and, in particular, that magnetic inhomogeneity promotes triplet pairing at S-F interfaces. Further support for this conclusion is provided by the observation of a temporal evolution of proximity-induced superconducting spectral features that resembles that of magnetic-structure-related transport effects in LCMO films. Our results have direct implications to the emerging field of superconducting spintronics [20] by showing that control over the magnetization configuration is a crucial factor in the design of corresponding devices.

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[1] L. N. Bulaevskii, A. I. Buzdin, and S. V. Panjukov, Solid State Commun. **44**, 539 (1982).

[2] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).

[3] R. S. Keizer, S. T. B. Goennenwein, T. M. Klapwijk, G. Miao, G. Xiao, and A. Gupta, Nature (London) **439**, 825 (2006).

[4] M. S. Anwar, F. Czeschka, M. Hesselberth, M. Porcu, and J. Aarts, Phys. Rev. B **82**, 100501 (2010).

[5] M. S. Anwar and J. Aarts, Supercond. Sci. Technol. **24**, 024016 (2011).

[6] D. Sprungmann, K. Westerholt, H. Zabel, M. Weides, and H. Kohlstedt, Phys. Rev. B **82**, 060505 (2010).

[7] T. S. Khaire, M. A. Khasawneh, W. P. Pratt, Jr., and N. O. Birge, Phys. Rev. Lett. **104**, 137002 (2010).

[8] J. W. A. Robinson, J. D. S. Witt, and M. G. Blamire, Science **329**, 59 (2010).

[9] C. Visani, Z. Sefrioui, J. Tornos, C. Leon, J. Briatico, M. Bibes, A. Barthelemy, J. Santamaria, and J. E. Villegas, Nat. Phys. **8**, 539 (2012).

[10] K. Dybko, K. Werner-Malento, P. Aleshkevych, M. Wojcik, M. Sawicki, and P. Przyslupski, Phys. Rev. B **80**, 144504 (2009).

[11] Y. Kalcheim, T. Kirzhner, G. Koren, and O. Millo, Phys. Rev. B **83**, 064510 (2011).

[12] Y. Kalcheim, O. Millo, M. Egilmez, J. W. A. Robinson, and M. G. Blamire, Phys. Rev. B **85**, 104504 (2012).

[13] Z. Sefrioui, D. Arias, V. Peña, J. E. Villegas, M. Varela, P. Prieto, C. León, J. L. Martinez, and J. Santamaria, Phys. Rev. B **67**, 214511 (2003).

[14] I. Fridman, L. Gunawan, G. A. Botton, and J. Y. T. Wei, Phys. Rev. B **84**, 104522 (2011).

[15] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.89.180506 for sample preparation information, the STM measurement process, and the temporal dependence of spectral features.

[16] A. F. Volkov and K. B. Efetov, Phys. Rev. Lett. **102**, 077002 (2009).

[17] M. Eschrig and T. Löfwander, Nat. Phys. **4**, 138 (2008).

[18] J. Linder, M. Cuoco, and A. Sudbø, Phys. Rev. B **81**, 174526 (2010).

[19] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. Lett. **86**, 4096 (2001).

[20] M. Eschrig, Phys. Today **64**(1), 43 (2011).

[21] E. C. Gingrich, P. Quarterman, Y. Wang, R. Loloee, W. P. Pratt, Jr., and N. O. Birge, Phys. Rev. B **86**, 224506 (2012).

[22] M. Ziese, S. P. Sena, and H. J. Blythe, J. Magn. Magn. Mater. **202**, 292 (1999).

[23] N. M. Nemes *et al.*, IEEE Trans. Magn. **44**, 2926 (2008).

[24] M. G. Blamire, B. S. Teo, J. H. Durrell, N. D. Mathur, Z. H. Barber, J. L. M. Driscoll, L. F. Cohen, and J. E. Evetts, J. Magn. Magn. Mater. **191**, 359 (1999).

[25] M. Houzet and A. I. Buzdin, Phys. Rev. B **76**, 060504 (2007).

[26] A. F. Volkov, F. S. Bergeret, and K. B. Efetov, Phys. Rev. Lett. **90**, 117006 (2003).

[27] J. Chakhalian *et al.*, Nat. Phys. **2**, 244 (2006).

[28] J. Aarts, S. Freisem, R. Hendrikx, and H. W. Zandbergen, Appl. Phys. Lett. **72**, 2975 (1998).

[29] R. Beck, Y. Dagan, A. Milner, A. Gerber, and G. Deutscher, Phys. Rev. B **69**, 144506 (2004).

[30] J. H. Ngai, R. Beck, G. Leibovitch, G. Deutscher, and J. Y. T. Wei, Phys. Rev. B **82**, 054505 (2010).