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# Long length scale interaction between magnetism and superconductivity in $La_{0.3}Ca_{0.7}MnO_3/YBa_2Cu_3O_7$ superlattices

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**Abstract.** We examine the interplay between ferromagnetism and superconductivity in $La_{0.3}Ca_{0.7}MnO_3/YBa_2Cu_3O_7$ superlattices showing "coexistence" of magnetism and superconductivity. A depression of the critical temperature is observed when manganite layer thickness is increased between 0.9 and 8 nm, pointing to an interplay between ferromagnetism and superconductivity over nanometer length scales. The possibility of a ferromagnetic/superconducting proximity effect to explain these results is analyzed.

**PACS.** 74.78.Fk Multilayers, superlattices, heterostructures – 74.25.Fy Transport properties (electric and thermal conductivity, thermoelectric effects, etc.)

## Introduction

The competition between magnetism and superconductivity has been the focus of considerable research effort in recent years [1–8]. The interplay between these two antagonistic long range orderings gives rise to quite exotic phenomena like spatial modulation of the order parameter, pi-junctions [9–13], etc, whose study, apart of its fundamental interest, may also open the way to important applications in the field of spintronics [14]. Ferromagnetic/superconductor superlattices based on high $T_c$ (HTS) and colossal magnetoresistance (CMR) materials offer a new scenario to explore this interplay at the nanometer scale. The spin polarization of the conduction band of the manganites is expected to suppress the superconductivity over very short length scales into the ferromagnet, and the short coherence length of the superconductor will make superconductivity to survive over very short length scales. Although there has been recently a theoretical effort to examine the F/S interface in oxides [15], to the best of our knowledge, experimental studies on the interplay between ferromagnetism and superconductivity are scarce in the literature. In this paper, we explore this issue on ferromagnetic/superconducting (F/S) $La_{0.3}Ca_{0.7}MnO_3$(LCMO)/ $YBa_2Cu_3O_7$ (YBCO) superlattices. The small lattice mismatch between LCMO and YBCO allows the growth of heterostructures with little structural disorder [16–19]. For this study we have grown superlattices with fixed YBCO thickness and changed systematically the thickness of the LCMO layer. Interestingly, magnetization (SQUID) and transport measurements show the "coexistence" of magnetism and superconductivity in superlattices with nanometer scale thickness of the individual layers. While the thinnest LCMO layers (3 unit cells) leaved the superconducting critical temperature almost unchanged, thicker LCMO layers result in a systematic reduction of the critical temperature over a wide thickness interval of the manganite layer. These results suggest an interplay between magnetism and superconductivity extending over long (few nanometers) length into the ferromagnet which is consistent with a F/S proximity effect.

## Experiment

We have grown LCMO/YBCO superlattices by high oxygen pressure (3.4 mbar) sputtering technique epitaxially on (100) $SrTiO_3$ (STO) at high temperatures (900 °C). YBCO single films on STO (100) were epitaxial with $T_c$ of 90 K and transition widths smaller than 0.5 K. LCMO single films, on the other hand, had ferromagnetic transition temperature, $T_{\rm CM}$, in excess of 200 K, and saturation magnetization $M_S$ = 400 emu/cm$^3$, close to the bulk values. Superlattices were grown positioning YBCO and LCMO targets alternatively over the heated substrate. Bottom layer was always LCMO since it grows better on STO

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than YBCO. Different sets of samples were grown with ferromagnetic and superconducting top layer.

## Results and discussion

We have grown superlattices with YBCO thickness fixed at 5 and 12 unit cells per bilayer and varied the thickness of the LCMO layer between 1 and 70 unit cells up to a total thickness of 150 nm. Samples were epitaxial and interfaces were atomically flat with little structural disorder as shown by X_ray diffraction analysis and transmission electron microscopy [18,19]. Quantitative analysis of the X-ray diffraction spectra shows no evidence for epitaxial strain and very limited roughness consisting in a fluctuation of layer thickness (step disorder) with an r.m.s. value of 0.19–0.27 nm (0.5–0.7 LCMO unit cells). Quantitative electron energy loss spectroscopy in a scanning transmission microscope did not show significant interdiffusion at the interfaces [20]. Further details about sample preparation and structure characterization can be found elsewhere [18–20].

Figure 1 shows hysteresis loops for superlattices with 8, 15 and 40 LCMO unit cells at 85 K, a temperature above the superconducting onset for these samples (see Fig. 2). Substantial depression of the sample magnetization is observed for manganite layer thickness below 15 unit cells (5.8 nm), although the samples were ferromagnetic even for the thinnest (3 unit cells) LCMO layers. Figure 2 shows resistance curves of superlattices with fixed YBCO thickness and changing the thickness of the magnetic layers. YBCO thickness is 5 unit cells in Figure 2a and 12 unit cells in Figure 2b. Resistance curves display sharp superconducting transitions, and show a systematic depression of the critical temperature when LCMO thickness is increased. The normal state resistance of the samples with thickest manganite layers show the metal insulator transition at the Curie temperature. Results on the variation of the critical temperature with magnetic layer thickness are collected in Figure 3 for superlattices with 5 (Fig. 3a) and 12 (Fig. 3b) unit cells thick YBCO layers. It can be observed that the critical temperature decreases in both cases down to a saturation value. Different data sets in Figures 3a and b correspond to samples with a superconducting (open symbols) and a ferromagnetic (solid symbols) top layer. The first result is that the length scale for superconductivity depression when the thickness of the magnetic layer is changed is different when the top layer is YBCO or when it is LCMO. This shows that part of the "action" is taking place into the YBCO, and most likely, this behavior is due to pair breaking by injected spin polarized carriers from the manganite. This mechanism would depress the critical temperature up to a length scale given by the spin diffusion length of the injected spin polarized carriers, which can be very long (several nanometers) above the superconducting gap [21]. Indeed, a depression of the critical current resulting from spin injection [21] has been observed before in YBCO/LCMO bilayers with thicker individual layers (50 nm). In this sense it is worthwhile to notice that the superlattices with a thick (12 u.c.)

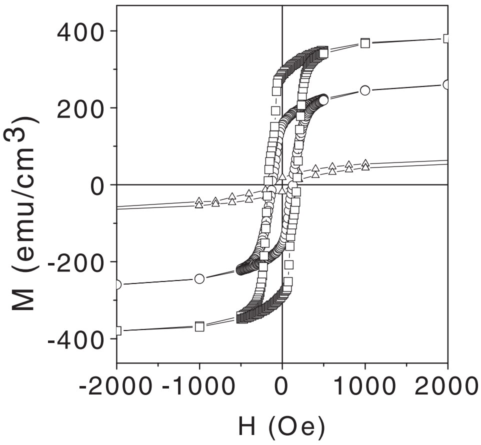


**Fig. 1.** Hysteresis loops at 85 K (above the superconducting onset) for superlattices with 12 unit cells thick YBCO. LCMO thickness is 8 unit cells (triangles), 15 unit cells (circles) and 40 unit cells (squares).

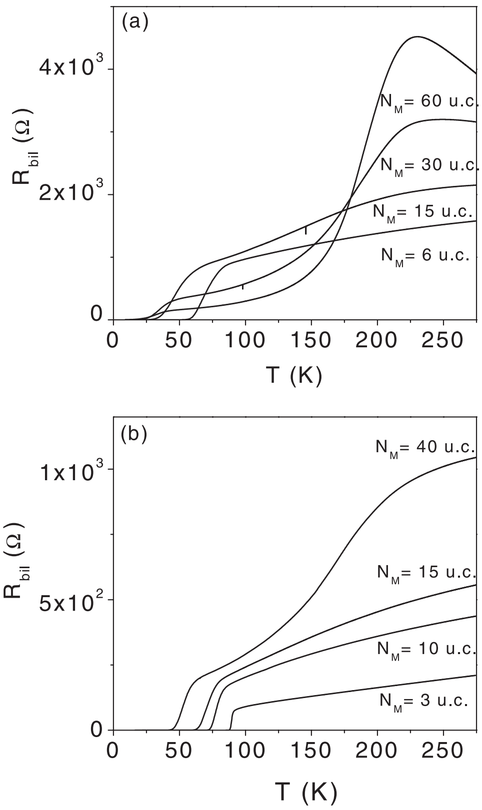


**Fig. 2.** Resistance curves of superlattices with a ferromagnetic top layer for constant YBCO layer thickness of 5 unit cells (Fig. 2a) and 12 unit cells (Fig. 2b). Thicknesses of the ferromagnetic layers are as labeled in the figure.

YBCO top layer do not exhibit a significant depression of the critical temperature showing that this thickness (14 nm) is an upper bound for the spin diffusion length. In addition, the latter result allows discarding disorder induced during growth as the possible explanation for the depression of the superconductivity. In second term, we will compare the results of samples with different YBCO thicknesses with ferromagnetic top layers (solid symbols

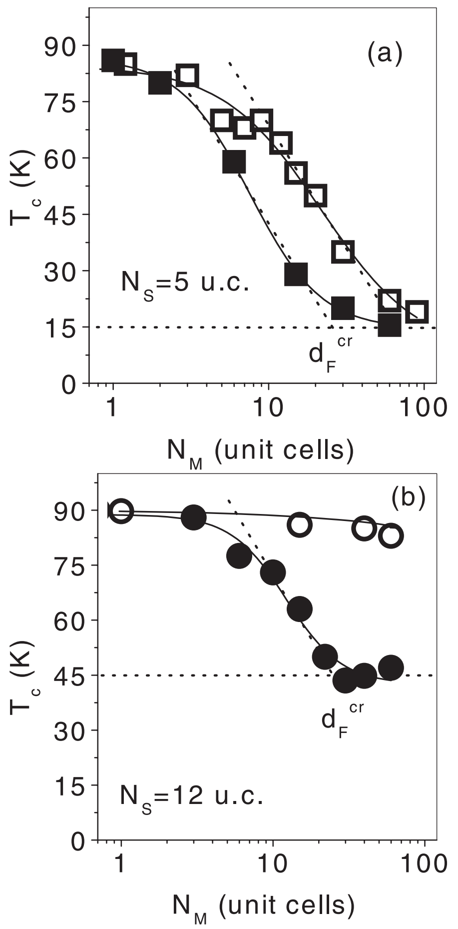


**Fig. 3.** Superconducting critical temperature as a function of magnetic layer thickness for superlattices with 5 (Fig. 3a) and 12 (Fig. 3b) unit cells thick YBCO layers. Different data sets in Figures 3a and b correspond to samples with a superconducting (open symbols) and a ferromagnetic (solid symbols) top layer. Continuous lines are a fits to a sigmoidal function. Dotted lines mark saturation temperature and the steepest slope of the decay to obtain the critical distance $d_F^{cr}$ (see text).

in Figs. 3a and 3b). We observe that although the critical temperature is depressed in both cases down to different saturation values the step like change occurs over the same LCMO thickness interval for both series of samples, suggesting a similar length scale for the depression of the superconductivity despite the different YBCO thicknesses. This length scale, $d_{cr}^F$, can be obtained by extrapolating the steepest slope in the transition curve to $T_c$ saturation level (see Fig. 3) and it is estimated to be $d_{cr}^F = 10$ nm for both series of samples. This is a very interesting result pointing to a common length scale for superconductivity depression into the ferromagnet and constitutes a clear indication of ferromagnetic/superconducting proximity effect. Cooper pairs entering into the ferromagnet experience the exchange interaction, which has a strong pair breaking effect and makes the order parameter to decay into the ferromagnet with a characteristic length scale $\xi_F$ [22]. Making the identification $d_{cr}^F = 2\xi_F$ (since there are magnetic layers at both sides of the superconducting layers) a value of $\xi_F$ is obtained of 5 nm. It is worth pointing that this value is much larger than expected according to the existing theories of the F/S proximity effect between a ferromagnet and an *s*-wave superconductor [22], which estimate $\xi_F = hv_F/\Delta E_{ex}$ (where $v_F$ is the Fermi velocity and $\Delta E_{ex}$ the exchange splitting). Given the large exchange splitting of the LCMO (3 eV) and a Fermi velocity for the majority band of $7.4 \times 10^7$ cm/s [23], the former expression yields very small values for $\xi_F$ of 0.2 nm (much smaller than the estimated 5 nm value). Moreover, the spin polarized nature of the conduction band of the LCMO is expected to weaken the F/S proximity effect if not completely suppress it [24]. However, we want to remark that we are in front of a very complex scenario not considered by the existing theories of the F/S proximity effect, in which the ferromagnet is fully spin polarized and the superconductor has an unconventional pairing symmetry (*d*-wave).

In ferromagnet/(*d*-wave) superconductor junctions with the interface perpendicular to the ab plane, the transmitted quasiparticles experience different signs of the pairing potential, which results in the formation of zero energy bound states (ZES) close to the surface [25] which are detected as zero energy peaks in tunnelling conductance spectra. In fact, theoretical reports on the tunnelling conductance in ferromagnet/*d*-wave superconductor double tunnel junctions show that ZES originate an enhancement of the quasiparticle tunnelling current [26]. Other ZES mediated processes like crossed Andreev reflection or elastic co-tunneling have been also reported very recently to enhance the conductance of F/S/F junctions [27]. Although it is clear that ZES may dominate transport properties of F/S/F structures, it is worthwhile to note that in the geometry of our experiment the interface is parallel to the ab plane (perpendicular to the *c*-direction) and it is not clear to which extent ZES could influence the F/S proximity effect in our samples, especially since there is not a theory for the proximity effect in ferromagnet/unconventional superconductor junctions.

Finally, a possibility that we cannot disregard is that the ultrathin manganite layer used in this work were not fully spin polarized. In fact, the reduced magnetization values found for the thinnest manganite layers in YBCO/LCMO superlattices could be reflecting reduced spin polarization levels. Spin polarization depends strongly on the state of the surface and phase separation or interface effects due to the small mismatch strain might influence the degree of spin polarization in our samples, thus allowing a longer rage F/S proximity effect according to well established theories [24].

In summary, we have found evidence for a long range interplay between ferromagnetism and superconductivity in oxide LCMO/YBCO superlattices. Our results point to the existence of a long range proximity effect in this system, although we can not discard the injection of spin polarized carriers into the superconductor as an additional source for superconductivity depression. Future work will be directed to clarify the relative importance of both mechanisms.

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