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# Magnetism and superconductivity in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superlattices

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We report on the magnetic and superconducting properties of  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/\text{YBa}_2\text{Cu}_3\text{O}_7$  (LCMO/YBCO) superlattices. For a constant LCMO layer thickness of 6 unit cells (u.c.), resistance and susceptibility measurements show superconductivity for YBCO layer thickness in excess of 4 unit cells. The critical temperature increases with YBCO thickness, and a  $T_c$  of 58 K is found for a YBCO thickness of 10 unit cells. Magnetization measurements show a ferromagnetic transition at 100 K in a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$  bilayer superlattice, and a depressed value of the saturation magnetization of  $20 \text{ emu cm}^{-3}$ . These results are discussed in terms of interface disorder (analyzed by x-ray diffraction and transmission electron microscopy) and of the possible interaction between magnetism and superconductivity. © 2001 American Institute of Physics.  
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## I. INTRODUCTION

Manganites with perovskite structure have been the focus of great interest in recent years due to its interesting magnetotransport properties and colossal magnetoresistance (CMR).<sup>1-5</sup> The oxide perovskite structure with pseudocubic lattice parameter of 0.386 nm of the  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  (LCMO) compound is quite adequate for the growth of heterostructures with high  $T_c$  superconducting cuprates, in particular  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (YBCO). This is interesting for both fundamental and applied studies. On one hand, this is an adequate system to study the interaction between magnetism and superconductivity, and the growth of heterostructures may be interesting for further understanding of tunneling phenomena; on the other hand, it opens new paths for the development of new tunneling magnetoresistance devices.

In this work we report on the *in situ* growth of (LCMO/YBCO) superlattices by high oxygen pressure dc sputtering on (001)  $\text{SrTiO}_3$  substrates. This technique combines high pressure gas atmosphere with high substrate temperature to yield low deposition rates ( $0.2 \text{ \AA/s}$ ), producing a very thermalized and ordered growth. We have previously shown the ability of this technique to growth epitaxial high  $T_c$  superlattices and trilayer tunnel junctions with  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) or  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  electrodes with sharp interfaces and a high degree of structural perfection.<sup>6,7</sup> For the purpose of this study we have fixed the LCMO thickness at 6 unit cells (u.c.), and varied the YBCO thickness between 3

and 10 u.c. The structure is analyzed by x-ray diffraction (XRD) and transmission electron microscopy (TEM). We show the presence of magnetism and superconductivity, although there is a depression both of the critical temperature of the superconductor and of the ferromagnetic transition temperature of the manganite.

## II. EXPERIMENT

Superlattices were produced *in situ* by sequential deposition of the films using a multitarget dc-sputtering system at high oxygen pressure on (100)  $\text{SrTiO}_3$  (STO) single crystal substrates. Stoichiometric sintered  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_{3-x}$  sputtering targets of 35 mm diameter were used. Pure oxygen at a pressure of 3.8 mbar was used as the sputtering gas, and a potential difference of 240 V with a current of 100 mA was applied between the electrodes. The substrate heater temperature was held at  $T_H = 900^\circ\text{C}$  during deposition. The structure was analyzed with high angle XRD using  $\text{CuK}\alpha$  radiation. The roughness of the individual layers was obtained by refinement using the SUPREX software.<sup>8</sup> TEM analysis was carried out using a JEOL 4000E $\times$  microscope operated at 400 kV. Cross-sectional samples for TEM were prepared by conventional mechanical grinding, dimpling, and argon ion milling with an acceleration voltage of 5 kV and an incidence angle of  $8^\circ$ . Magnetotransport measurements were conducted in a He cryostat with a 8 T superconducting magnet, and magnetization was analyzed using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design).

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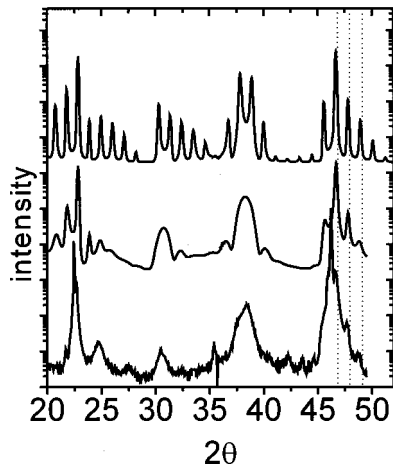


FIG. 1. High angle XRD pattern of a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$  bilayer superlattice. The upper curve is a SUPREX simulation with perfect layers. The middle curve is a fit with 2 unit cell fluctuation in the LCMO layer thickness and 0.8 unit cell fluctuation in the YBCO layer thickness. The lower curve is the experimental XRD pattern. Vertical dashed lines mark superlattice peaks.

This deposition procedure consistently produces epitaxial  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_{3-x}$  thin films with good structural properties as proven by several analytical techniques such as XRD, TEM, and atomic force microscopy (AFM) analysis. We optimized the growth conditions to produce single films with optimal properties for both materials. YBCO films had critical temperatures in excess of 90 K with a wide transition of 0.1 K, and showed good epitaxial properties. Details on the quality of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films have been reported elsewhere.<sup>6,7</sup> LCMO films also grew epitaxially, with a metal-insulator transition of  $T_{M-I} = 282$  K, a Curie temperature of  $T_c = 270$  K, and a saturation magnetization of  $400 \text{ emu/cm}^3$  at 10 K.

### III. RESULTS AND DISCUSSION

Figure 1 shows a typical high angle XRD pattern of a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$ . Since the pseudocubic lattice parameter of the manganite is about one third of the  $c$  lattice parameter of the YBCO superlattice, Bragg peaks nearly occur at Bragg peaks of the individual layers. Sharp superlattice satellites can be seen around the manganite peaks yielding a modulation length of 7 nm for the superlattice. Superlattice peaks are marked with vertical dashed lines in Fig. 1. The YBCO peaks, on the other hand, are wide, reflecting a significant amount of roughness in the manganite layer breaking the coherence between the YBCO layers. Simulations of the XRD pattern using the SUPREX software yielded a roughness of the manganite layer (step disorder) of 2 unit cells, and about 1 unit cell for the YBCO layer. Figure 2 shows a cross-sectional low magnification image of the  $(\text{LCMO}_6/\text{YBCO}_5)$  sample with the electron beam perpendicular to the multilayer  $c$  axis. A clear contrast between the superconducting and magnetic layer is observed, the layers being continuous over lateral distances on the order of thousands of angstroms. This is at variance with recent reports of noncontinuous spiral like growth on STO substrates.<sup>9</sup> How-

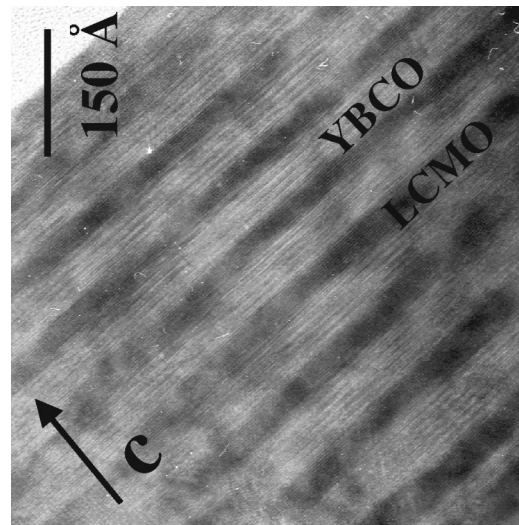


FIG. 2. TEM image of a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$  bilayer superlattice. The thickness fluctuation of the LCMO layers can be readily seen.

ever, the interfaces are not perfectly flat, showing clear layer thickness fluctuations at long length scales (hundreds of angstroms). High magnification images (not shown) confirm the presence of interface steps of one or two manganite unit cells high (4–8 Å). Such observations also confirmed the coherent character of interfaces, provided the small in plane lattice mismatch between YBCO and LCMO, i.e., at the short length scales probed by HREM the interfaces are atomically flat (between steps). It is important to remark that, from the two structural probes used in this study (XRD and TEM), we cannot definitely exclude chemical interdiffusion between both layers. Refined x-ray patterns accounted reasonably well for experimental data without the presence of interdiffusion, although 10% interdiffusion in the interface layer did not produce significant changes in the confidence factor of the fit ( $\chi^2$ ).

Samples were superconducting (established from resistivity and from mutual inductance measurements) for YBCO thickness larger than 4 unit cells  $[(\text{LCMO}_6/\text{YBCO}_N)_{15}]$ , with  $N > 4$ . Figure 3 displays the resistance curves of a  $(\text{LCMO}_6/\text{YBCO}_5)_{15}$  superlattice showing a zero resistance

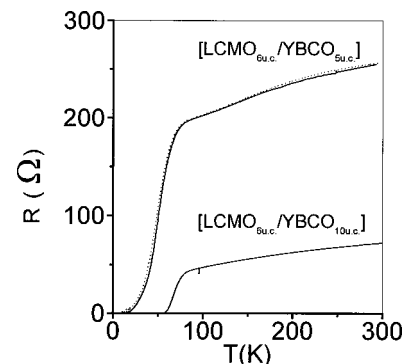


FIG. 3. Upper curve: Resistance curves of a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$  bilayer superlattice with zero applied field (solid line), and with  $H = 40 \text{ kOe}$  applied parallel to the substrate (dashed line). Lower curve: Resistance curves of a  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{10 \text{ u.c.}})_{15}$  bilayer superlattice with zero applied field.

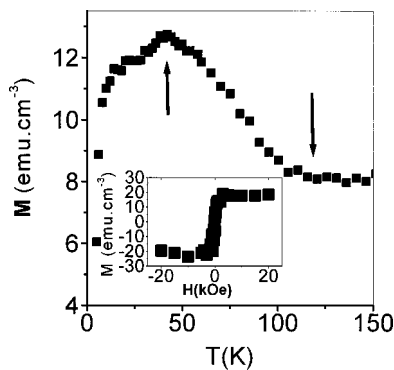


FIG. 4. Magnetic moment vs temperature of  $(\text{LCMO}_{6 \text{ u.c.}}/\text{YBCO}_{5 \text{ u.c.}})_{15}$  bilayer with an applied field of  $H=2 \text{ kOe}$ . Arrows indicate the ferromagnetic transition ( $T=100 \text{ K}$ ) and the onset of the superconducting transition ( $T=50 \text{ K}$ ). The inset shows the magnetization loop at  $T=50 \text{ K}$ .

transition temperature of 20 K, and of a  $[\text{LCMO}_6/\text{YBCO}_{10}]_{15}$  sample with a  $T_c$  of 58 K. The resistance curves did not show the ‘‘bump’’ characteristic of the metal–insulator transition at the ferromagnetic transition. This reflects that the manganite layer shows enhanced resistivity due to interface disorder, and that current flows through the YBCO layers. This effect is observed in YBCO/PBCO superlattices, in which current basically flows through YBCO layers below 250 K.<sup>6</sup> This is not completely unexpected in our superlattices, since an increase of the low temperature resistivity has been observed previously in strained layers grown on  $\text{LaAlO}_3$  and  $\text{NdGaO}_3$ .<sup>9</sup> In fact, samples with a YBCO thickness smaller than 4 unit cells  $[(\text{LCMO}_6/\text{YBCO}_N)_{15}, \text{ with } N < 4]$  were insulating and showed much higher values of the normal state resistivity (2000  $\Omega$  at room temperature). Interestingly, resistance curves of the  $(\text{LCMO}_6/\text{YBCO}_5)_{15}$  superlattices do not show transition broadening upon the application of a magnetic field up to 4 T parallel to the substrate (dashed line in Fig. 3). This has been previously observed in two-dimensional (2D) like, 1 unit cell YBCO thick layers in YBCO/PBCO superlattices with spacer thickness in excess of 5 unit cells, and results from the decoupling of  $\text{CuO}$  plane blocks by the thick PBCO layers.<sup>6,10</sup> In our case, this behavior could arise from the suppression of superconductivity down to a thickness comparable with one unit cell.

Measurements of the sample magnetic moment as a function temperature (see Fig. 4) show a clear ferromagnetic transition at about 100 K. At lower temperatures (below 50 K), the signature of the diamagnetic response of the YBCO layers shows up, reducing the total magnetic moment of the sample, which becomes negative at lower temperatures. Magnetization loops at 50 K (above the superconducting transition temperature) show a clear, but small, magnetic moment, and a saturation field of 2 kOe (see the inset in Fig. 4). The saturation magnetization,  $M_s$ , obtained from the hysteresis loops was  $20 \text{ emu cm}^{-3}$  on the basis of 15 LCMO layers each 6 unit cells thick. This value is much lower than the one found in single films ( $400 \text{ emu cm}^{-3}$  at 10 K). This can partly arise from the high temperature of the measurement in relation to the ferromagnetic transition temperature.

Additionally, reduced ferromagnetic transition temperatures and small values of the magnetic moment have been previously observed in thin single layers and LCMO/STO superlattices,<sup>11</sup> and are explained in terms of epitaxial strain and/or dead layers arising from surface or interface disorder.<sup>9,12,13</sup> In fact, in view of the 2 unit cell roughness obtained from x-ray analysis substantial layer thickness fluctuation can cause local changes of the magnetic moment. This points to the value of  $20 \text{ emu cm}^{-3}$  being somewhat underestimated. Additionally, although single films showed optimum *in situ* oxygenation, with ferromagnetic transition temperatures close to 300 K, there is the possibility of deficient oxygenation of the superlattices due to the layered nature of the samples. Finally, we cannot exclude the magnetism being depressed by the presence of the superconducting layer. In fact a depression of the magnetic properties due to the presence of superconducting layers has been previously reported in metallic superlattices.<sup>14,15</sup>

A last remark with regard to the low value of the superconducting critical temperature. X-ray analysis with the SUPREX software showed roughness values (layer thickness fluctuation) of the YBCO layers of 0.8 unit cells. Even if we assume that one complete unit cell at the interface would not be superconducting, the critical temperature of  $(\text{LCMO}_6/\text{YBCO}_5)_{15}$  superlattices is still too low (20 K) compared, for example, with YBCO/PBCO superlattices. We have previously shown that 3 unit cell YBCO layers in YBCO/PBCO superlattices have a  $T_c$  in excess of 70 K.<sup>6</sup> Moreover, the  $T_c$  of 58 K of a  $(\text{LCMO}_6/\text{YBCO}_{10})_{15}$  sample can hardly be explained in terms of interface properties. However, we cannot exclude deficient oxygenation of the layers to some extent limited by slower oxygenation dynamics of the manganite layers (a  $T_c$  of 58 K would correspond to an oxygen content of 6.6 per formula for single films). Another interesting possibility is that the reduced  $T_c$  in our magnetic/superconducting multilayers might also be a fingerprint of the suppression of the superconductivity by the magnetic layers in this system.<sup>14,15</sup>

#### IV. CONCLUSIONS

In summary, we have presented artificially layered materials, showing magnetism and superconductivity. The superconducting and ferromagnetic critical temperatures are depressed. Further work is necessary to clarify to what extent this depression arises from interface disorder and deficient oxygenation or from the interaction between magnetism and superconductivity.

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