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Magnetism and Superconductivity in $Nd_{0.81}Sr_{0.19}MnO_3/YBa_2Cu_3O_7$ Superlattices

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We report on the growth, structural and magnetic characterization of $Nd_{0.81}Sr_{0.19}MnO_3/YBa_2Cu_3O_7$ (NSMO/YBCO) superlattices. The NSMO system for the doping level of $x = 0.19$ is a ferromagnetic insulator. Multilayers with a fixed NSMO thickness of 13 unit cells and a varying YBCO layer thickness from 2 unit cells to 6 unit cells were sputtered on $LaAlO_3$ substrates. An onset of superconducting transition is seen starting from the multilayer with 3 unit cells of YBCO layer thickness. Hysteresis loops recorded above and below the superconducting transition show a signature of interlayer exchange coupling.

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1. Introduction

Multilayered structures allow materials to have different physical properties and long-range ordering to be placed into close proximity. The interaction between the layers gives rise to new phenomena. Experiments on ferromagnetic metal/normal metal (F/N) multilayers [1] show an indirect exchange coupling between F layers via N layers. An open question is whether an indirect exchange coupling between ferromagnetic (F) layers across superconducting (S) layers can be transferred.

A recently published theoretical model [2] suggests that an indirect exchange coupling between F layer through S layer is possible The appearance of a superconducting gap in the spacer introduces a new length scale. The range of such interaction is lower than 13 nm. The indirect exchange coupling is thought to be oscillatory both above and below the superconducting critical temperature. According to the author, the most relevant heterostructure to study such an effect are multilayers composed of colossal magnetoresistance materials (CMR) and high temperature superconductors (HTSC).

The doping level for manganese perovskites controls the ground state of the system. According to the phase diagram [3] of $Nd_{1-x}Sr_xMnO_3$ system when $x < 0.22$ the system is a ferromagnetic insulator.

Our motivation in the present study is to gather information about the signature of the interlayer indirect exchange coupling between ferromagnetic insulating NSMO layers across *d*-wave superconducting $YBa_2Cu_3O_7$ spacing layers.

2. Experimental

$Nd_{0.81}Sr_{0.19}MnO_3/YBa_2Cu_3O_7$ (NSMO/YBCO) multilayers were deposited on (100) $LaAlO_3$ substrates using sequential high-pressure dc sputtering [4]. The sputtered multilayers were deposited at 770°C in 3 mb of oxygen pressure. Two targets with nominal composition of $Nd_{0.81}Sr_{0.19}MnO_3$ and $YBa_2Cu_3O_7$ were used for deposition. The thickness of different layers was controlled by the deposition times of the respective targets. The calibrated deposition rates were 1.3 nm and 2.6 nm per minute for NSMO and YBCO, respectively, X-ray diffraction measurements and cross-section transmission microscope verified the superlattice structure. Magnetisation versus temperature, $M-T$, and magnetic field M versus H, measured magnetic properties using a SQUID magnetometer. Samples used for SQUID measurements were 3×3 mm^2 in size. Resistance versus temperature measurements were performed with a four-probe method.

3. Results and discussion

We have deposited $[\mathrm{NSMO} \times 13\ \mathrm{u.c.}/\mathrm{YBCO} \times n\ \mathrm{u.c.}]_{16}$ multilayers, in which the NSMO layers thickness were fixed at 13 unit cells (u.c.) and the YBCO layer thickness was varied from 1 unit cell to 6 unit cells. The *c*-axis lattice parameter for relaxed YBCO film is 1.168 nm. The NSMO 100 nm layer thickness film deposited on $LaAlO_3$ substrate has a lattice parameter of about 0.391 nm, whereas for the bulk polycrystalline $Nd_{0.81}Sr_{0.19}MnO_3$ sample a is 0.386 nm. To determine the quality of multilayers grown by our method we have prepared and structurally characterized NSMO/YBCO superlattices. High angle diffraction data revealed the presence of modulation satellites. The obtained modulation period within 10% was in agreement with the nominal modulation length, demonstrating the accuracy of the thickness calibration. This result was also confirmed by TEM cross-section studies. Figure 1 shows a cross-section TEM micrograph of the $[\mathrm{NSMO} \times 10\ \mathrm{u.c.}/\mathrm{YBCO} \times 4\ \mathrm{u.c.}]_{16}$ sample, in which different layers can by clearly distinguished, because of a high atomic number contrast between the layers. It is

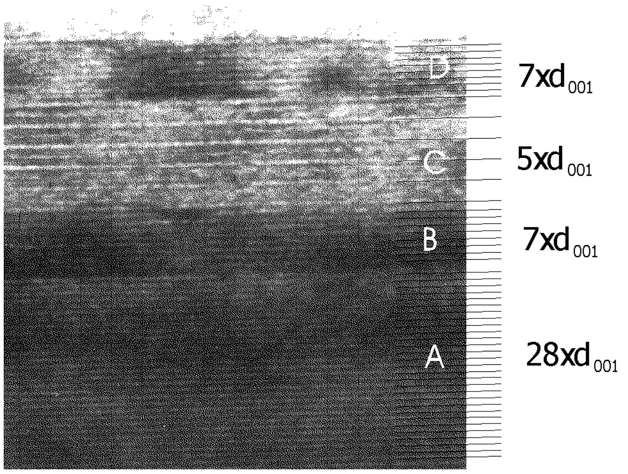

Fig. 1. TEM cross-section image of the [NSMO × 10 u.c./YBCO × 4 u.c.]$_{16}$ multilayer.

seen that the two oxides grow heteroepitaxially parallel to each other; however, a thickness fluctuation of about 1 unit cell is seen.

Figure 1 shows also a high-resolution fringes image of the region covering $LaAlO_3$ substrate, NSMO first layer, YBCO layer, and second NSMO layer.

It is seen that spacing between the planes of the first NSMO layer is larger than for the second NSMO layer by about 10%. Based on the lattice parameter of the $LaAlO_3$ substrate $a = 0.379$ nm we have determined the 001 plane spacing for the first NSMO layer equal to 0.41 nm. This observation shows that the first NSMO layer plays a role of buffer layer. Figure 2a shows resistance versus temperature relation $R(T)$ of measured multilayers (Table). Starting from the [NSMO × 13 u.c./YBCO × 3 u.c.]$_{16}$ sample a transition to superconducting state is observed.

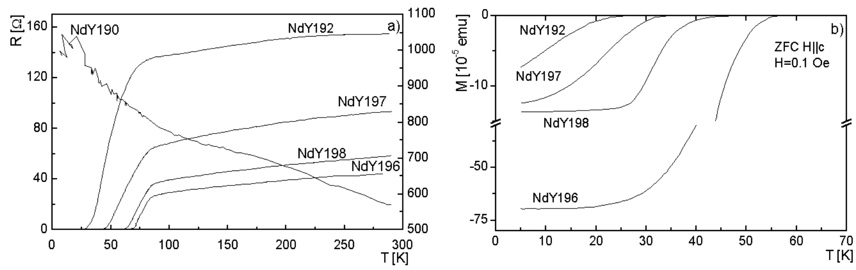

Fig. 2. Resistance versus temperature of NSMO/YBCO superlattices (a); ZFC magnetic moment vs. temperature for NSMO/YBCO superlattices (Table) (b).

Also in $La_{0.67}Ca_{0.33}MnO_3/YBa_2Cu_3O_7$ (LCMO/YBCO) superlattices a transition to superconducting state is observed beginning with the multilayer with 3 unit cells of YBCO layers thickness [5].

TABLE

Modulation length and zero resistance temperature of the NSMO/YBCO multilayers.

Sample No	Nominal modulation length	T_{c0} [K]
NdY196	$[Nd_{0.81}Sr_{0.19}MnO_3(5.2\ nm)/YBCO(7.2\ nm)]_{16}$	61
NdY198	$[Nd_{0.81}Sr_{0.19}MnO_3(5.2\ nm)/YBCO(6.0\ nm)]_{16}$	56
NdY197	$[Nd_{0.81}Sr_{0.19}MnO_3(5.2\ nm)/YBCO(4.8\ nm)]_{16}$	40
NdY192	$[Nd_{0.81}Sr_{0.19}MnO_3(5.2\ nm)/YBCO(3.6\ nm)]_{16}$	25
NdY190	$[Nd_{0.81}Sr_{0.19}MnO_3(5.2\ nm)/YBCO(2.4\ nm)]_{16}$	–

A sample NdY190, i.e. with 2 unit cells of YBCO layer thickness, demonstrates a semiconducting-like behaviour of $R(T)$. Such behaviour could be related to lattice distortion of both subsystems or to interdiffusion at NSMO/YBCO interface.

Figure 2b shows zero field cooling (ZFC) magnetic moment versus temperature of measured superlattices (Table). The onset of diamagnetic response temperature T_{d0} corresponds to the zero resistance temperature T_{c0}. For $Nd_{0.67}Sr_{0.33}MnO_3$/YBCO superlattices [6] we observed a lower diamagnetic response temperature than the zero resistance transition temperature.

In Fig. 3 we present a field cooled (FC) magnetic moment of the single $Nd_{0.81}Sr_{0.19}MnO_3$ thin film. It is seen that the Curie temperature is about 100 K. ZFC and FC magnetic moments versus temperature recorded for the [NSMO × 13 u.c./YBCO × 3 u.c.$]_{16}$ superlattice demonstrate that the Curie temperature for the sample is also of about 100 K, however, for this multilayer a paramagnetic background is observed. The inset in Fig. 3 shows resistance versus temperature relation indicating an insulating state at temperatures below 150 K.

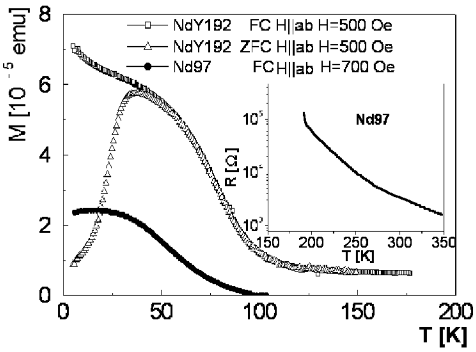

Fig. 3. Field cooling magnetic moment for the NSMO single film, Nd97, and ZFC, and FC magnetic moment of the $[Nd_{0.81}Sr_{0.19}MnO_3 \times 13\ u.c./YBCO \times 3\ u.c.]_{16}$, NdY192, multilayer. The inset shows resistance vs. temperature dependence of the NSMO, Nd97, single film.

In Fig. 4 we present hysteresis loops of studied multilayers. The $M-H$ curves were recorded in the 10 K to 60 K temperature range for magnetic field parallel ($H \parallel ab$) and perpendicular to substrates ($H \parallel c$). As it is presented in Fig. 4a–f hysteresis loops recorded at a temperature of $T = 10$ K for $H \parallel c$ show a response from YBCO layers. In consecutive multilayers, with an increase in YBCO layer thickness the irreversibility of magnetic moment, ΔM_{irr}, is increasing. $M-H$ curves recorded at 60 K present a response from NSMO layers.

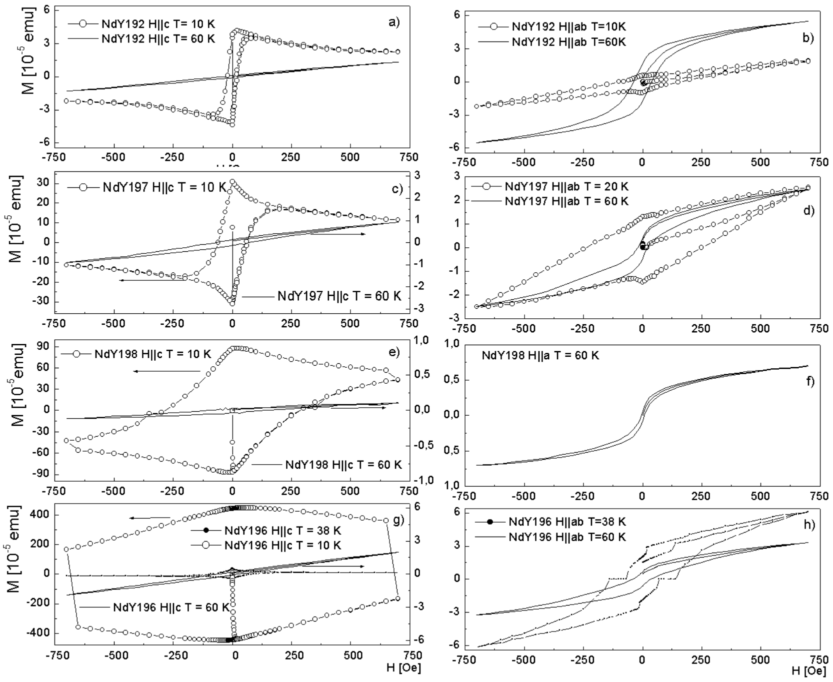

Fig. 4. 4 Left panels: Magnetization loops of the NSMO/YBCO multilayers Table) measured at 10 K to 60 K temperature range for $H \parallel c$; right panels: magnetization loops of the NSMO/YBCO multilayers (Table) measured at 10 K to 60 K temperature range for $H \parallel ab$.

Hysteresis loops recorded for magnetic field parallel to the substrates are presented on the right panel of Fig. 4. The hysteresis loop measured for the [NSMO × 13 u.c./YBCO × 3 u.c.]$_{16}$, NdY192, sample presents a superposition of magnetic moment of NSMO layers and magnetic moment from superconducting YBCO layers.

It is also clear that because of large shape anisotropy of studied multilayers the in-plane [1 0 0] direction is the easy axis of magnetic moment. On the other hand, for magnetic field perpendicular to the multilayers, i.e. [0 0 1] direction, the hysteresis loops have a shape characteristic of hard axes. This observation indicates that any interlayer exchange coupling in NSMO/YBCO superlattices is related to in-plane rotation of magnetic moments in consecutive NSMO layers, i.e. the ground state oscillates from antialignment (AF) to alignment (F) magnetic moments of NSMO layers.

$M-H$ curves measured at a temperature of 60 K demonstrate an oscillation in the shape of the hysteresis loops, which is indicative of the existence of interlayer coupling [7] between the magnetic layers across YBCO spacing layers. On the other hand, the observation of plateau on $M-H$ curve (Fig. 4h) demonstrates an antiparallel alignment of magnetic moments in consecutive NSMO layers in a multilayered structure.

In summary, we have grown high-quality NSMO/YBCO superlattices. TEM studies indicate that superlattices have a well-defined superlattice structure. An occurrence of superconductivity is observed beginning with the sample with 3 unit cells of YBCO layer thickness. Hysteresis loops indicate a presence of interlayer exchange coupling between NSMO layers across YBCO spacer layers.

Acknowledgments

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References

[1] P. Grünberg, R. Schreiber, Y. Pang, M.B. Brodsky, H. Sowers, *Phys. Rev. Lett.* **57,** 2442 (1986).

[2] C.A.R. Sa de Melo, *Physica C* **387,** 17 (2003).

[3] Y. Tokura, Y. Tomioka, *J. Magn. Magn. Mater.* **200,** 1 (1999).

[4] P. Przyslupski, A. Wisniewski, R. Szymczak, J. Igalson, *Czech. J. Phys.* **46,** 1355 (1996); P. Przyslupski, S. Kolesnik, E. Dynowska, T. Skoskiewicz, M. Sawicki, *IEEE Trans. Appl Supercond.* **7,** 2192 (1997).

[5] P. Prieto, P. Vivas, G. Campillo, E. Baca, L.F. Castro, M. Varela, C. Ballesteros, J.E. Villegas, D. Arias, C. León, J. Santamaría, *J. Appl. Phys.* **89,** 8026 (2001).

[6] I. Komissarov, E. Dynowska, M. Sawicki, A. Szewczyk, P. Przysłupski, *Phys. Status Solidi A* **196,** 66 (2003); P. Przysłupski, I. Komissarov, P. Dłużewski, J. Pełka, E. Dynowska, M. Sawicki, *Physica C* **387,** 40 (2003).

[7] A. Orozco, S.B. Ogale, Y.H. Li, P. Fournier, Eric Li, H. Asano, V. Smolyaninova, R.L. Green, R.P. Sharma, R. Ramesh, T. Venkatesan, *Phys. Rev. Lett.* **83,** 1680 (1999).