Single crystal substrates effect on the critical behavior in La$_{2/3}$Ca$_{1/3}$MnO$_3$ thin films

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Abstract. In this work we study the substrate influence in magnetic properties and the critical behavior of La$_{2/3}$Ca$_{1/3}$MnO$_3$ thin films grown on single-crystal substrate of LaAlO$_3$, SrTiO$_3$ and NdGaO$_3$, using a DC-magnetron sputtering system. All films were growth under the same conditions and with thicknesses close to 130 nm. We made measures of magnetization as a function of temperature between 150 and 300 K, around critical temperature (Tc) with different external applied magnetic fields (100 Oe – 30 kOe). X rays measures were made to verify the existence of La$_{2/3}$Ca$_{1/3}$MnO$_3$ (LCMO) phase, where we observed two peaks corresponding to the samples with Miller index (020) and (040) that verify the presence of LCMO phase in these thin films. Furthermore, critical exponent ($\beta$) was obtained fitting the magnetic measures with a proposed model. $\beta$ values for each substrate are around 0.25, 0.27 and 0.36, that show an important influence of substrate.

1. Introduction
Manganites have been matter of study in recent years because they have presented the phenomenon of colossal magnetoresistance (CMR) which is promising for future technological applications, in particular the Manganites family of type La$_{1-x}$AxMnO$_3$ (A = Ca, Sr, Ba, Pb) [1]. This paper presents a study of critical magnetic behavior of the magnetization as a function of temperature in the vicinity of the magnetic transition zone for La$_{2/3}$Ca$_{1/3}$MnO$_3$ (LCMO) thin films, grown on single crystalline substrates of LaAlO$_3$ (LAO), SrTiO$_3$ (STO) and NdGaO$_3$ (NGO). The analysis of how the type of substrate influence on the critical exponents is done by characterizing the critical behavior (magnetic transition Ferro-Paramagnetic) of this compound.

2. Experimental procedure
LCMO thin films were grown on LaAlO$_3$ (LAO), SrTiO$_3$ (STO) and NdGaO$_3$ (NGO) single-crystalline substrates (100) under the same conditions deposition by using DC-magnetron sputtering system. During the deposition the substrate temperature was kept at 850 °C, the pressure of high purity oxygen was 500 mTorr, the substrate-target distance was 40 mm and the deposition power was 35 W. The X-ray diffraction measurements were performed with a Bruker D8 Advance, with a step of 0.02° and 3 s per step. Magnetic measurements were performed via vibrating-sample magnetometer technique (VSM) with the physical properties measurement system (PPMS, Quantum Design). All measurements were taken by using a 40-Hz vibration frequency for the detection coil with 2-mm The magnetization vs. temperature M vs. T after zero field cooling (ZFC) was measured with an applied external magnetic field H of 300 Oe to 30 kOe.
3. Results and discussion

After obtaining the films became the structural analysis by X-ray measurements. Figure 1 shows X-ray diffraction patterns associated with LCMO/LAO, LCMO/STO and LCMO/NGO thin films. All films are single phase without any detectable impurity or secondary phase. Figure 1 (a), shows that the most intense peaks correspond to substrates; the reflections in 23.0° and 47.06° with Miller indice (020) and (040), respectively, correspond to LCMO phase grown on LAO and STO, respectively. However, films grown on NGO present only one peak that belongs to LCMO associated to Miller indice (040) due to orthorhombic crystalline structure of both, by that reason the sample peaks are shielded by substrate peaks. All LCMO peaks on different substrates were fitted using Gaussian and Lorentzian functions. In the figure 1 (b), we show how looks the fit for one film grown on LAO. The lattice parameters were calculated with the peaks positions found from the fits. The values were $a = 5.4215(3)$ Å, $b = 5.4325(2)$ Å and $c = 7.6855 (3)$ Å.

Regarding the magnetic properties, in the figure 2 are shown the $M$ vs. $T$ curves taken in each of the samples. LCMO/LAO and LCMO/STO measurements exhibit the typical magnetic behavior of this compound. The change in the magnetic transition is not abrupt, i.e, magnetization is not zero in a precise value of temperature as is expected for an ideal ferromagnetic material. In this case there is a range of temperature where magnetization is becoming zero, this is attributed to the magnetic inhomogeneous nature of the samples. Such behavior is more evident with high magnetic fields. Furthermore, magnetization values are strongly influenced by the kind of using substrate. Films grown on STO have bigger magnetization values than those grown on LAO due to the coupling between the structure and each substrate. On the other hand, in films grown on NGO submitted to high magnetic fields (between $H= 6$ and 30 kOe), shows a different behavior of the others, the magnetic transition slope decreases as the $H$ increases until the transition disappears. However, typical magnetic transition of LCMO maganites is observed at lower fields of 1000 kOe (see inset figure 2. (c)).

In order to analyze the critic nature of magnetic moment as a function of temperature around the transition region, the proposed model in [2] was used. Figure 2 (d), shows the fits obtained for each sample with external magnetic field of 1000 Oe, which are in excellent agreement with experimental data. Figure 3 (a), exhibit values of critical exponent $\beta$ vs $H$ given by the fit. For samples grown on LAO and STO, $\beta$ varies in a non-monotonically for smaller field, with values between 0.14 and 0.25. Whereas that, for large fields $> 4000$ Oe, $\beta$ values do not change considerably, these fluctuate between 0.24 and 0.27. This indicates that the critical magnetic behavior of the samples does not depend strongly on the substrates in this range of magnetic fields. However, in NGO samples the critical

![Figure 1](image1.jpg)

**Figure 1.** (a) XRD patterns of LCMO thin films on LAO, STO and NGO. (b) Open circles are experimental data and the continuous lines are the fits on LCMO/LAO.
The exponent $\beta$ takes values between 0.28 and 0.36, (see inset figure 3 (a)) which are greater than the substrates previous, the highest value is related with magnetic behavior type Heisenberg [3].

Extrapolating the $\beta$ values from figure 3 (a) y (b), we found the $\beta$ (H=0), these values are shown in table 1. LCMO/STO and LCMO/LAO films have $\beta$ values of 0.27 and 0.26 respectively, these values are not related to a particular kind of universality, however we consider this result interesting because these values are typical of systems that exhibiting “tricritical” point values, which have been found in these same manganites systems [4]. As to magnetic transition width ($\Delta T$) as a function of $H$ were also obtained from the fits. As shown in figure 3 (b), we divide the $\Delta T$ behavior in regions (1) and (2). In region (1) $\Delta T$ does not vary with $H$ for each of the substrates. Nevertheless in region (2) for $H>4000$ Oe, $\Delta T$ values tend to increase linearly with the applied field, in this case $\Delta T_c$ has contributions from both the magnetic inhomogeneity and the applied magnetic field. It is important to note that $\Delta T$ follows a power law type behavior as a function of $H$ of the form $\Delta T = \Delta T_c + c H^{\eta/\delta}$ [2], where $\eta$ is a critical exponent associated with the dependence of the magnetization with $H$ around the $T_c$. $\Delta T_c$ is the width of transition to $H = 0$ and $c$ is a constant dependent of the sample. $\eta$ values were obtained from $\Delta T (H)$ fit using the above expression which are shown in table 1. For LCMO/LAO samples these values are nearly to Heisenberg type behavior, but for STO and NGO the found values are lower. Assuming the relation taken from [2] between the exponents $\eta = \beta \delta$ values were calculated for each substrate which are between 3 and 7.

Figure 2. (a), (b), (c) Magnetic moment as a function of temperature. (d) Fit, circles are the experimental data and the solid line is the fit.
Figure 3. (a) $\beta$ exponent as a function of applied external magnetic field in LCMO thin films, (b) $\Delta T_C$ as a function of applied external magnetic field in LCMO thin films, the zone (1) and (2) are visual guides. The inset shows Polynomial fit on $\Delta T$ values for the LCMO/STO film. Squares are experimental data and line is the fit.

Table 1. Values for $\beta (H=0)$, $\eta$ and $\delta$.

| Substrate | $\beta (H=0)$ | $\eta$     | $\delta$    |
|-----------|--------------|------------|-------------|
| SrTiO$_3$ (STO) | $0.27 \pm 0.01$ | $1.34 \pm 0.12$ | $5.0 \pm 0.6$ |
| LaAlO$_3$ (LAO)  | $0.26 \pm 0.01$ | $1.72 \pm 0.20$ | $6.6 \pm 0.8$ |
| NdGaO$_3$ (NGO)  | $0.36 \pm 0.01$ | $1.21 \pm 0.40$ | $3.4 \pm 1.2$ |

4. Conclusions

In summary, in this work LCMO films were grown on STO, LAO and NGO substrates. The magnetic measurements show a large dependence with the substrate used. Different behavior was observed in the analysis done in LCMO/NGO films. $\beta$ values obtained were 0.32 and 0.36 indicating that the magnetic behavior of this system is related to the Heisenberg model 3-D.

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