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Possible influence of the ferromagnetic/antiferromagnetic interface on the effective critical behavior of bilayers based on $La_{1-x}Sr_xMnO_3$

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ABSTRACT

In this work, the effective critical exponent of the spontaneous magnetization, β, and the transition temperature, T_C, were calculated from magnetization measurements of three bilayers based on $La_{1-x}Sr_xMnO_3$ (LSMO). The bilayers structure is a ferromagnetic (FM) LSMO film grown on top of an antiferromagnetic (AF) LSMO film. The value of the antiferromagnetic film thickness was kept the same for the three samples, while the ferromagnetic film had different thickness for each bilayer. Applying a method of calculation based on a linear superposition of the magnetization close to the critical temperature, a β value corresponding to the 3D Ising model was found for the bilayer with the thinnest ferromagnetic film. This result, and the other obtained values are explained taking into account the possible influence of the FM/AF interface on the magnetic and crystal orderings.

1. Introduction

The manganites have been widely studied in the last years among other reasons, for their transport properties as colossal magnetoresistance, half-metallicity and as a possible choice for reading and writing devices [1]. The $La_{1-x}Sr_xMnO_3$has some advantages respect to the other mixed-valance manganites as its high Curie temperature, $T_C \sim 370$ K (bulk) and being the most metallic [2].

On the other hand, the study of the critical exponents associated to the ferromagnetic–paramagnetic transition can shed a light on the dimensionality, range of the interaction and symmetry of the order parameter (magnetization) [3] in this manganite. In fact, different values for the critical exponents of LSMO have been reported until now, varying according to the type of the sample (polycrystalline, single crystal, film) and the Sr doping. Ghosh et al. [4] found a critical exponent β of the temperature dependence of the spontaneous magnetization, equal 0.37 ± 0.04 for a single crystal with Sr-doping of $x=0.3$. Kim et al. [2] reported a similar value, $\beta=0.40 \pm 0.02$, also for a single crystal but with slightly different doping of 0.25. With even smaller $x=0.125$, Nair et al. [5] found $\beta=0.37 \pm 0.02$. In their summary of the critical exponents, they point out the difference among the values reported for LSMO single crystals with $x=0.3$, depending on the measuring technique. For instance, the β value by Ghosh et al. [4] was obtained by measuring bulk magnetization. Meanwhile, a sample with the same characteristic but using microwave absorption gave a $\beta=0.45 \pm 0.05$ [5].

When it comes to polycrystalline LSMO, Mohan et al. [6] determined a $\beta=0.50 \pm 0.02$ for $x=0.2$. Thus, the role that dimensionality and the symmetry play on the nature of the transition is evident.

In this work, the effective critical exponent β of three LSMO bilayers is determined by a method based on a linear superposition of different parts of the sample, each with its own critical temperature [7]. Since the thickness of the upper ferromagnetic layer is the parameter that changes from bilayer to bilayer, the purpose of this paper is to get some understanding of the influence, if any, of the FM/AF interface on the critical behavior of epitaxial LSMO thin films.

2. Material and methods

Three bilayers made of ferromagnetic LSMO ($x=0.35$) on antiferromagnetic LSMO ($x=0.65$) deposited on (001) $SrTiO_3$, STO, i.e. LSMO(FM)/LSMO(AF)/STO, were fabricated by ozone assisted molecular beam epitaxy, MBE (see Table 1). The substrate temperature and the working pressure were typically $T_s=750$ °C and

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Table 1
LSMO bilayers and thin film studied in this work. The substrate is STO for all samples.

Sample (unit cells)	Description
1. Bilayer 25/100	LSMO(FM)/LSMO(AF)
2. Bilayer 50/100	LSMO(FM)/LSMO(AF)
3. Bilayer 100/100	LSMO(FM)/LSMO(AF)
4. Film 40	LSMO(FM)

$PO_3 = 1 \times 10^{-6}$ mbar. The thickness of the AF-LSMO was around 38.0 nm (nominally 100 uc) for all bilayers, while the values of the thickness of the FM-LSMO film were close to 9.5 nm (25 uc), 19.0 nm (50 uc) and 38.0 nm (100 uc). The thickness error is around 10%, as determined by X-ray reflection.

The temperature dependence of the magnetization for each sample was measured by vibrating sample magnetometry (VSM, Versalab Quantum Design), from 50 K to 370 K. The sample was cooled down with an applied field of 100 Oe, then the field was switched off and the curve was taken while warming. In this way, a truly or intrinsic width of the temperature transition, ΔT_C, can be determined, avoiding the field-broadening effect [7].

From the measured M vs. T curves, the effective critical exponent for the spontaneous magnetization, β, and the critical temperature T_C of the phase transition were calculated. This was done by the method of Berger et.al. [7]. It is based on the fact that magnetization vs. temperature curve presents a "tail", thus indicating a range of possible values for T_C. Applying the theory of superposition, it is assumed that the curve can be adjusted by least squares, using the equation

$$M_{(T)} = \frac{m_0}{\sqrt{2\pi}\Delta T_c}\int_{Tc}\left(\frac{T_c - T}{T_c}\right)^{\beta}\theta(T_c - T)\exp\left[\frac{-1}{2}\left(\frac{T_c - T_{c,avg}}{\Delta T_c}\right)^2\right]dT_c \quad (1)$$

where β is the effective critical exponent governing the transition and ΔT_c represents the standard deviation of the critical temperature range. This value is proportional to the width of the interval, and therefore, the degree of inhomogeneity of the material. $T_{c,avg}$ is the average value of the critical temperature, T_c, being the most frequent value within the sample.

The method was used due to its accuracy, since when adjusting, this equation perfectly fits to the data [7,8]. Furthermore, by using this model, $La_{0.67}Ca_{0.33}MnO_3$ thin films have shown a tail in the magnetization vs. temperature curve, indicating a strong dependence of the type of substrate in the inhomogeneity of epitaxial films [9], as is well known in manganites [10–13]. Therefore, these facts were used to study the FM/AF interface, by analyzing the values of β, $T_{c,avg}$ and ΔT_c parameters.

Besides, the magnetization of a LSMO film ($x = 0.35$) 40 uc thick also grown on STO was measured as well (Table 1), but with an applied field of 500 Oe. This broadens the width of the transition but it should not affect the obtained value of the β exponent. In fact, the eq. 1 is derived taking β as field-independent [8]. This assumption seems to break down when the sample is non-epitaxial and polycrystalline [8], but all the samples studied here are epitaxial MBE-grown films.

3. Results and discussion

The normalized magnetization vs. T curves for the three bilayers are presented in Fig. 1. By applying the method described in the previous section, the magnetization was adjusted according to Eq. (1) for the samples indicated in Table 1.

The fit of the graph of magnetization in e.m.u vs. temperature in Kelvin, is shown in Fig. 2. The data are in blue circles, and the fitted curve is in red. As it can be seen there is an excellent fit therein. Therefore, the parameters obtained are very reliable. The values obtained for the effective critical exponents and T_C are shown in Table 2.

According to these results, the two bilayers with the thinner FM layer (25 uc and 50 uc) have very similar transition temperature and width values. It is important at this point to notice also that both their magnetization curves show a characteristic "*dip*" at a temperature around 220–230 K. Taking into account the reported Neel temperature for LSMO single crystals (without applied field) and polycrystals being around 230 K [14,15], it makes sense that the "*dip*" can be a signature of the AF transition of the bottom film of the bilayer. These similarities could be related to some influence of the interface: since the layer below is AF some kind of death magnetic layer at the interface FM/AF can be presented, affecting the temperature and width of the ferromagnetic to paramagnetic transition. These three features are characteristic for these two samples but not for the bilayer with thicker (100 uc) FM film.

On the other hand, when it comes to β, the two closer values are for the bilayers with ferromagnetic layers 50 uc and 100 uc thick. Meanwhile, the bilayer with the geometry 25 uc/100 uc gave the value $\beta = 0.325 \pm 0.003$, corresponding to the 3D, $n = 1$, Ising model [3]. This can be due to the presence of a preferential axis of magnetization or easy axis, while for the other two bilayers, being thicker, this tendency is smeared out. This easy axis can be induced due to some interface coupling to the AF layer. This was deduced mainly due to the thickness of the AF-LSMO was the same for the three samples (100 uc), as well as the substrate. Further, the values of the critical exponents depend on the spatial dimensionality of the sample, d, the order parameter dimensionality, n, and the range of the exchange interaction $J(r)$ [16]. Thus, for the three bilayers d was the same, and hence $J(r)$ and n could vary when the influence of the interface FM/AF becomes important (Mainly because of the thickness relation changes between the samples).

The obtained values of β for the structures with unit cells 50/100 and 100/100, do not correspond to any of the universality class for second order phase transition, and they lie in a range between the 3D theories and the mean field one. As known, this critical exponent also depends on the range of the interaction. Thus, for these two bilayers, a possible slower decaying with the distance of the exchange interaction takes place in contrast to the 25/100 bilayer.

Meanwhile, for the single film a value of $\beta = 0.495 \pm 0.001$ (Table 2) was obtained, corresponding to the mean field theory, indicating that the exchange interactions are of long-range type. This value is consistent whit the results obtained by Khiem et.al. [16] for $La_{0.7}Sr_{0.3}Mn_{0.8}Ti_{0.2}O_3$, and confirm the conclusion of Mohan et.al. [6], that mean field interactions could be important in manganites.

Lofland et al. [17] also reported a mean field behavior ($\beta = 0.45 \pm 0.05$) for a $La_{0.7}Sr_{0.3}MnO_3$ single crystal, measured by three techniques using microwave absorption. The theoretical explanation of these behaviors is the following: The values of the β critical exponent depend on d, n, and $J(r)$. This function was modeled by Fisher et.al. [18] as $J(r) = 1/r^{\sigma + d}$, were, if $\sigma \leq 1/2$, the renormalization group analysis predict the mean field critical exponent.

The apparent non-correlation of the β and ΔT_c values can be explained as following:

On one hand, the transition width ΔT_c is related to the presence of inhomogeneities [7] which in this case, taking into