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Structural and magnetic properties of La$_{0.7}$Sr$_{0.3}$MnO$_3$ thin films integrated onto Si(100) substrates with SrTiO$_3$ as buffer layer

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La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO), which belongs to the family of the perovskitelike manganites, is expected to present a spin polarization close to 100% (half metal). This material has been therefore used in a number of spin-dependent transport devices grown on insulating and matching substrates [SrTiO$_3$ (STO) and NdGaO$_3$] such as magnetic tunneling junctions and spin valves. Moreover, LSMO is a good candidate for spin injection into semiconductors, due to its low carrier density, its high spin polarization of charge carriers, and its ferromagnetism at the room temperature. Therefore, the integration of LSMO as a source of spin polarized carriers in semiconductor based devices is a challenging task for potential applications utilizing both information processing and data storage in the same device, such as high-density magnetic memories and magnetic sensors.

In order to grow an epitaxial quality manganite film on a semiconductor substrate, the film–substrate lattice mismatch needs to be minimized and chemical reactions between the substrate and the deposited film should be eliminated. The first attempts in order to obtain the epitaxial LSMO films on Si used a buffer intermediate layer consisting of STO or of Yttria stabilized zirconia (YSZ) while other ones took advantage of the SiO$_x$ native oxide film on the Si surface. Due to the recent progresses concerning the direct integration of STO on Si, the STO buffer layers are now often for integrating LSMO onto this technologically important semiconductor. Up to now, the intense research activity on LSMO thin films grown on Si substrates was mainly focused on their structural and static magnetic properties, but according to our knowledge their dynamic magnetic properties remain unexplored. Therefore, the aim of this paper is to investigate static and dynamic magnetic properties of LSMO thin films of various thicknesses deposited on a Si substrate.

LSMO films (with a thickness $d$ of 10, 20, 60, and 100 nm) were grown on Si (001) substrates by a reactive molecular beam epitaxy. For all samples, x-ray diffraction (XRD) revealed an excellent epitaxy with in-plane cubic [100] and [010] axes of LSMO. The XRD measured values of the out-of-plane lattice parameter suggest that the strain state does not vary significantly from sample to sample. A super conducting quantum interference device reveals that the room temperature magnetization at saturation increases with $d$ and nearly reaches the bulk value for $d = 100$ nm; the Curie temperature ranges in the 320–350 K interval, to compare to 360 K in the bulk. Ferromagnetic resonance (FMR) in cavity (at 9.5 GHz) and microstrip FMR used to investigate the dynamic magnetic properties, revealed a fourfold anisotropy showing its easy axes along the [110] and [110] directions. In the thickest samples ($d > 20$ nm), the FMR spectra present two distinct resonant modes. This splitting is presumably due to the simultaneous presence of two different magnetic phases. © 2011 American Institute of Physics.

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As previously published,\textsuperscript{15} the dynamic properties are tentatively interpreted assuming a magnetic energy density which, in addition to Zeeman and demagnetizing contributions, contains a perpendicular and an in-plane anisotropy contribution

\begin{equation}
E = -M_s H [\sin \theta_M \sin \theta_H \cos(\varphi_M - \varphi_H) + \cos \theta_M \cos \theta_H]
- (2 \pi M_s^2 - K_\perp) \sin^2 \theta_M
- \frac{1}{8} K_4 (3 + \cos 4(\varphi_M - \varphi_H)) \sin^4 \theta_M
\end{equation}

As detailed in Ref. 15, \( \theta_M \) and \( \varphi_M \), respectively, represent the out-of-plane angle and the in-plane angle (referring to the [100] axis) defining the direction of the magnetization \( M_s \); \( \varphi_H \) stands for the angle between a fourfold easy axis with the [100] direction. \( K_4 \) is chosen positive with this definition. As in Ref. 15 we introduce the effective magnetization field

\[ 4\pi M_{\text{eff}} = 4\pi M_s - 2K_4/M_s = 4\pi M_s - H_\perp \]

and the fourfold in-plane anisotropy field \( H_4 = 4K_4/M_s \).

The \( H \) in-plane and \( H \) out-of-plane MS-FMR spectra of all the samples reveal a single uniform precession mode, except for the 100-nm-thick film for which an additional mode is observed. In this last sample, the lower and the higher frequency modes are called mode 1 and mode 2, respectively: With an in-plane applied field, their frequency difference increases with the direction normal to the sample plane (and large enough to allow for \( M_s/H \)). The measured frequencies linearly vary versus \( H \), as expected from the studied model [Fig. 2(b)]. In the 100-nm-thick film, the frequency difference between mode 1 and mode 2 is independent of \( H \).

Additional investigations relative to the temperature dependence of the resonance spectra were performed using conventional FMR. Depending on the temperature and on the studied film, these spectra show one or two uniform modes: In the 100-nm-thick sample, the occurrence of two modes is restricted to the 240–300 K interval, while, for the 60-nm-thick film, such a multiplicity only advents in the 120–200 K region and, for the other samples, is always absent. These differences are not yet fully understood. Anyway, a critical analysis of the experimental results clearly argues for the uniform character of both modes when the two above mentioned distinct resonances are observed.

The four studied samples present very similar structural and magnetic characteristics, except for the above mentioned multiplicity appearing in the thickest films (60 and 100 nm). In the following the experimental results concerning these last two samples which show an unusually richer behavior are presented and expected frequency variations versus the applied field. Such typical dependences are shown on Fig. 2(b). The Landé factor is found independent of the sample geometry. These differences are presumably related to the interfacial strain through the magneto-elastic coupling. It can be easily proved that, due to the order of magnitude of the magnetic parameters and of the available applied fields, the in-plane geometry does not allow for a precise simultaneous determination of \( g \) and of the effective demagnetizing field \( 4\pi M_{\text{eff}} \) from the variation of the resonance frequency versus the magnitude of the applied field. Such typical dependences are shown on Fig. 2(b). The Landé factor is found independent of the sample \( (g = 1.915) \) but \( 4\pi M_{\text{eff}} \) substantially varies: The deduced values of \( 4\pi M_{\text{eff}} \) are reported in Table I.

The MS-FMR technique, in perpendicular configuration, allows for deriving the values of the effective Landé factor \( g \) and of the effective demagnetizing field \( 4\pi M_{\text{eff}} \) from the variation of the resonance frequency versus the magnitude of the applied field. Such typical dependences are shown on Fig. 2(b). The Landé factor is found independent of the sample \( (g = 1.915) \) but \( 4\pi M_{\text{eff}} \) substantially varies: The deduced values of \( 4\pi M_{\text{eff}} \) are reported in Table I.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|}
\hline
\textbf{Thickness (nm)} & \textbf{10 nm} & \textbf{20 nm} & \textbf{60 nm} & \textbf{100 nm} \\
\hline
\textbf{Mode 1} & \textbf{Mode 2} & \textbf{Mode 1} & \textbf{Mode 2} & \textbf{Mode 1} & \textbf{Mode 2} \\
\hline
\textbf{4\pi M_s (kOe)} & 2.4 & 2.8 & 2.8 & 4 & 5.6 \\
\textbf{4\pi M_{\text{eff}} (kOe)} & n.m. & 4.9 & 4.9 & 4.7 & 5.6 \\
\hline
\textbf{4\pi M_{\text{eff}} (kOe)} & 4.6 & 4.9 & 4.9 & 5 & 6 \\
\textbf{H_x (kOe)} & 50 & 8 & 28 & 30 & 38 \\
\hline
\end{tabular}
\caption{Magnetic parameters at room temperature obtained from the best fits to our experimental results (n.m.: not measured). \( H_x \) is calculated using the \( 4\pi M_{\text{eff}} \) values derived from in-plane measurements.}
\end{table}
magnetic fields in order to put in evidence rather weak magnetic anisotropies which cannot be detected using the high applied magnetic fields required in conventional FMR. Taking an advantage of this opportunity, we derived the in-plane anisotropy at the room temperature of all the samples from the resonance frequency variations versus the direction (defined by $\phi_H$) of an in-plane applied field of a small fixed amplitude. Figure 2(c) shows such typical experimental angular dependences in the 100-nm-thick sample. For all the investigated films the experimental data are satisfactorily fitted using the model described in Ref. 15 on the basis of Eq. (1). In all the cases we find: $\phi_4 = 45^\circ$, which means that the [110] (and, indeed, the [100]) direction defines a principal axis, as expected from our XRD study and, moreover, that it corresponds to an easy direction. The values derived for $H_d$ are reported in Table I. Notice that in the 100-nm, $H_d$ gets different values in mode 1 and mode 2. This is confirmed by additional measurements at lower temperature using conventional FMR. This result confirms the hypothesis that both modes are uniform modes with distinct effective demagnetizing fields and distinct anisotropies and excludes, for instance, the identification of mode 2 to a perpendicular standing excitation. Consequently, we think that, at least in some temperature and thickness ranges, two different magnetic phases can coexist and that they define distinct uniform modes showing different effective demagnetizing and anisotropy fields.

The occurrence of two modes in the thickest samples prevents for deriving unambiguous conclusions concerning the influence of the thickness $d$ on the magnetic parameters. However, as illustrated on Table I, the magnetization $M_s$ tends to decrease when $d$ decreases, as generally observed, and the anisotropy fields tend to increase, which argue for their interfacial character inducing a partial relaxation along a direction normal to the film and, consequently, their enhanced effective values when $d$ is reduced. A quantitative model, taking in account the energy terms related to the interfacial strains evoked in Ref. 15, is presumably too ambitious, regarding to the observed dispersion of the experimental results.

The temperature dependence of the static and dynamic magnetic properties of 10, 20, 60 and 100-nm-thick LSMO films, deposited on Si substrates overlaid by a 20 nm STO buffer layer have been studied. Our x-ray diffraction measurements show an excellent epitaxial (001) orientation of the LSMO films. The magnetic behavior is interpreted assuming a magnetic energy density characterized by a strong uniaxial anisotropy along the direction normal to the plane of the films and by an in-plane anisotropy showing a fourfold symmetry. The four studied samples present very similar structural and magnetic characteristics, except for the unusual presence of two uniform magnetic resonance modes in the thickest samples. This mode multiplicity arises from the presence of two distinct magnetic phases.

FIG. 2. (Color online) Field dependence of the resonance frequency of the uniform excited modes of the 60- and 100-nm-thick LSMO films. The magnetic field is applied (a) in-plane and (b) perpendicular to the film plane. (c) In-plane angular dependence of the resonance frequency of the uniform modes of the 100-nm-thick LSMO film. Fits are obtained using the parameters indicated in the Table I.