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Channelling study of $La_{1-x}Sr_xCoO_3$ films on different substrates

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ABSTRACT

The cobalt oxide system $LaCoO_3$ and its Sr-doped child compounds have been intensively studied for decades due to their intriguing magnetic and electronic properties. Preparing thin $La_{1-x}Sr_xCoO_3$ (LSCO) films on different substrates allows for studies with a new type of perturbation, as the films are subject to substrate-dependent epitaxial strain. By choosing a proper substrate for a thin film grow, not only compressing but also tensile strain can be applied. The consequences for the fundamental physical properties are dramatic: while compressed films are metallic, as the bulk material, films under tensile strain become insulating. The goal of this work is to determine the strain tensor in LSCO films prepared on $LaAlO_3$ and $SrTiO_3$ substrates by pulsed laser deposition using RBS/channelling methods. Apart from the composition and defect structure of the samples, the depth dependence of the strain tensor, the cell parameters, and the volume of the unit cell are also determined. Asymmetric behaviour of the strained cell parameters is found on both substrates. This asymmetry is rather weak in the case of LSCO film grown on $LaAlO_3$, while stronger on $SrTiO_3$ substrate. The strain is more effective at the interface, some relaxation can be observed near to the surface.

1. Introduction

$LaCoO_3$ perovskite and its Sr-doped derivatives have been subject of intensive studies for decades due to their intriguing magnetic and electronic properties that lead to a complex phase diagram including spin-state transition, colossal magnetoresistance and a cooperative ferromagnet – spin-glass and metal–insulator transition [1–9]. Despite all efforts, the complete understanding of the underlying physics has not been achieved yet.

One of the most important parameters to alter bulk physical properties is the effective Co–O–Co bonding angle, which determines the electronic and magnetic interactions between the cobalt and oxygen ions. This angle depends on the lattice parameters, which are usually altered by hydrostatic or chemical pressure. Preparing thin $La_{1-x}Sr_xCoO_3$ films on different substrates allows for studies with a new type of perturbation, as the films are subject to well controlled substrate-dependent epitaxial strain. By choosing a proper substrate for a thin film grow, not only compressing but also tensile strain can be applied. The consequences for the fundamental physical properties are dramatic: while compressed LSCO films are metallic, films under tensile strain, with otherwise unchanged parameters such as chemical composition become insulating [10]. Moreover, strain can induce ferromagnetism in the otherwise paramagnetic bulk $LaCoO_3$ material [11,12]. A few percent of difference in strain leads to many orders of magnitude difference in resistivity and magnetic interactions. This type of tunability of the electronic and magnetic properties by external parameters makes those thin film materials rather interesting for the emerging field of oxide-based electronics.

In the last decades, strain engineering has become an important and necessary technique to improve advanced nanoelectronic devices [13]. Apart from various X-ray [14], neutron [15] and electron [16] diffraction techniques, a powerful method for the non-destructive characterization of strain states in thin layer systems is Rutherford backscattering spectrometry combined with channelling effect (RBS/C) [17,18].

In this work channelling study have been performed on $La_{1-x}Sr_xCoO_3$ films prepared on $LaAlO_3$ and $SrTiO_3$ substrates to determine either the various types of defects and strain in the films.

2. Strain determination from channelling experiments

Strains in crystals are associated with changes in the angles between different crystal directions – except for direction conserving purely hydrostatic strain, i.e., dilatation or compression. From channelling measurements, only the "deviatory" part, $\tilde{\varepsilon}$, defined as

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the difference between the full strain tensor ε and its hydrostatic part, can be determined, i.e.,

$$\tilde{\varepsilon} = \varepsilon - \mathrm{trace}(\varepsilon/3)\cdot\mathbf{1} = \begin{pmatrix} (2\varepsilon_{11}-\varepsilon_{22}-\varepsilon_{33})/3 & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{12} & (2\varepsilon_{22}-\varepsilon_{11}-\varepsilon_{33})/3 & \varepsilon_{23} \\ \varepsilon_{13} & \varepsilon_{23} & (2\varepsilon_{33}-\varepsilon_{11}-\varepsilon_{22})/3 \end{pmatrix} \quad (1)$$

where **1** is the diagonal unit tensor. The trace of $\tilde{\varepsilon}$ vanishes by construction, i.e., the diagonal elements of $\tilde{\varepsilon}$ are not linearly independent of each other. In cubic system, the strain tensor components, ε_{ij} can be expressed with the changes in the angle between the direction of [001] and [h,k,l], δ_{hkl} (in radian) in the following way [18]:

$$\begin{aligned} &\varepsilon_{11}-\varepsilon_{33} = \delta_{101} + \delta_{\bar{1}01};\\ &\varepsilon_{13} = (\delta_{\bar{1}01} - \delta_{101})/2;\\ &\varepsilon_{22}-\varepsilon_{33} = \delta_{011} + \delta_{0\bar{1}1};\\ &\varepsilon_{23} = (\delta_{0\bar{1}1} - \delta_{011})/2 \text{ and}\\ &\varepsilon_{12} = \tfrac{3}{\sqrt{8}}(\delta_{111} - \delta_{\bar{1}11}) \end{aligned} \quad (2)$$

Let us note that H. Trinkaus and his co-workers presented an excellent paper [18] where they gave all the required relations between the strain induced angle changes and the components of the strain tensor for general crystalline layer systems of reduced symmetry compared to the basic (cubic) crystal.

3. Experimental

$La_{0.7}Sr_{0.3}CoO_3$ films were grown on single crystalline substrates of $SrTiO_3$ (001) (lattice parameter $a = 3.901$ Å) and $LaAlO_3$ (001) ($a = 3.789$ Å) substrates with a size of 5×5 mm^2, by pulsed laser deposition (KrF 248 nm) from a stoichiometric target. The deposition temperature and the oxygen background pressure were 650 °C and 3.5×10^{-1} mbar, respectively. The films were cooled down in 600 mbar of oxygen.

In order to perform proper orientation of the crystals using RBS/C a small part of the film (~1 mm from the edge of the sample) was removed by ion sputtering using 1 keV Ar using high incident angle of 80°. The glancing angle of incidence is necessary to avoid the defect accumulation, while for reducing the atomic mixing low ion energy is to be used [19].

Composition, defect structure and strain tensor were determined by RBS combined with channelling technique. RBS/C analysis was performed using the 5 MV Van de Graaff accelerator at the Institute for Particle and Nuclear Physics, Wigner Research Centre for Physics, Hungarian Academy of Sciences.

The samples were fastened to a sample holder of a scattering chamber containing a two-axes goniometer. During the experiments the vacuum in the chamber was better than 1×10^{-4} Pa using liquid N_2 traps along the beam path and around the sample. The ion beam of $^4He^+$ was collimated with 2 sets of four-sector slits to the necessary dimensions of 0.5×0.5 mm^2. An ion current of typically 10 nA was measured by a transmission Faraday cup [20]. The measured spectra were collected with a dose of 4 μC. The RBS measurements were performed with an ORTEC detector with a solid angle of 4.15 msr. The energy calibration of the multichannel analyzer was determined using known peaks and edges of Au, Si and C.

To determine the sample composition 2000 keV He RBS was performed at tilt angles of 7°, 45° and 56°, respectively. In order to avoid the channelling, the samples were rotated during the measurements. To study the defect structure of the samples RBS/channelling experiments in channel <001> were used with beam energies of 1500, 2000, 2500 keV. The elemental composition and the defect structures of the samples were evaluated from the

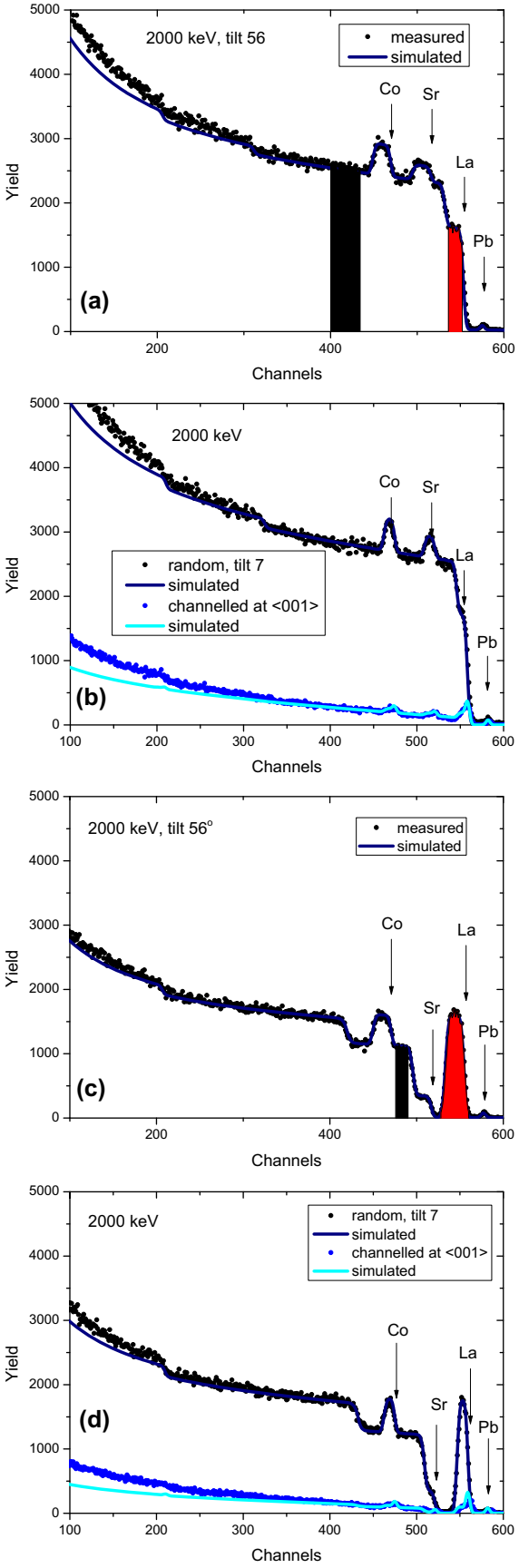

Fig. 1. Measured and simulated RBS (a and c) and RBS/channelling (b and d) spectra using beam energy of 2000 keV taken on $La_{1-x}Sr_xCoO_3$ $x = 0.3$ film grown on $LaAlO_3$ (a and b) and $SrTiO_3$ (c and d) substrates.

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