Modeling Of Residual Stresses In Thin Films Of Y_2O_3 Deposited On Si Substrate, $SrTiO_3$ AND M_gO

Abdellah Boualam*, Brahim Boubeker*, Abdellah Zamma*, Fabien Paumier**

**Laboratoire de Caractérisation des Matériaux, Faculté des Sciences Ben M'Sik Av. Driss Harti, Sidi Othman, Casablanca Morocco.*
***Université de Poitiers, UFR Sciences, S.P.2M.I, Laboratoire de Métallurgie Physique, URA 131 du CNRS, Bd 3, Téléport 2, BP. 179, 86960 Futuroscope cedex, France.*

Abstract: The study concerns the analysis of residual stress in thin films obtained by IBAD (Ion Beam Assisted Deposition). Only the thermal stress in the film can be calculated and it has no other information on other types of constraints. Kamminga and Al have proposed a model in which they describe the stress state as a combination of a hydrostatic stress caused by the introduction of particles into the holes of the matrix of the film and a fixing constraint to keep the lateral dimensions of the film identical to those of the substrate. We apply this model to determine the stresses in the films of yttrium deposited by IBS of Si substrates, SrTiO3 and MgO.

Keywords: Residual stress – Hydrostatic constraint– Fixing constraint – Biaxial constraint.

I. Introduction

Part of a range of materials with remarkable peculiarity, modifiable by extern elements (development, equilibrium with the outside atmosphere), yttrium oxide has a wide range of physical properties, superconductivity, electronic conduction, ionic conduction, high resistivity, magnetoresistance, laser proper-ties, thermal barrier, radiation resistance) and many potential applications. It is therefore essential to monitor the state of stress and their origins in this new generation of materials with complex microstructure

II. Caractérisation

II-1. Characterization of Y2O3 films

The experimental characterization combines the x-ray diffraction (XRD), the transmission electron microscopy (TEM) and backscat-tering spectroscopy (RBS)..

II-2. Sample preparation

Before each deposition, we must proceed to:

- The cleaning substrates with argon jet to remove dust surface;
- The insertion of substrates in the deposition chamber;
- The pumping to a pressure of about 10^{-8} Tor
- The heating the substrate ;
- The stripping the target of Y_2O_3 with three successive stages of 10 min (40, 60 and 80mA)

The diffraction pattern obtained in θ-2θ mode shows a single orientation of growth plans of the yttrium oxide as a function of substrate.

- ❖ On Si and MgO substrate: growth occurs along the [111] direction wich has a centered cubic structure;
- ❖ On $SrTiO_3$ substrate: Growth of fluorite type.

III. Modeling of stress state

The X-ray diffraction is very effective for measuring stress in thin films because it can probe the elastic response of crystalline materials to internal and external stresses

III-1. Sin²ψ method

The state of deformation in thin films is obtained by the sin²ψ method for measuring the lattice parameter (defor-med mesh) in different crystallographic directions. This can be traced back to the state of associated stress.
for this, we consider three systems of coordinates:

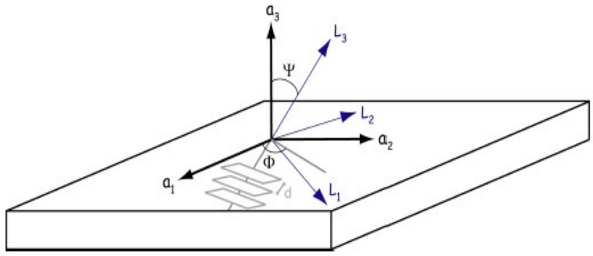

Figure 1: Landmark of sample and laboratory

- ➢ $\left(\vec{S}_1 \quad \vec{S}_2 \quad \vec{S}_3\right)$ is the reference sample such as $\vec{S}_3$ is oriented along the normal to the surface of the sample,

- $(\vec{a}_1 \ \ \vec{a}_2 \ \ \vec{a}_3)$ the crystallographic reference such as $\vec{a}_3$ is oriented along the normal to the crystallographic plane
- $(\vec{L}_1 \ \ \vec{L}_2 \ \ \vec{L}_3)$ the laboratory system such as $\vec{L}_3 = \vec{a}_3$ and deducted of the refernce sample S by:
  - ❖A first rotation in the plane of the sample at an angle Φ around the a_3;
  - ❖A second rotation around $\vec{S}_2 = \vec{L}_2$ at an angle ψ in order to make $\vec{L}_1$ per-pendicular to the crystallographic plane.

III-2. Hooke's Law

It connects the strain tensor and stress through the tensor compliances

$$\varepsilon_{ij} = s_{ijkl}\sigma_{kl}$$

Wich becomes with the Voigt notation

$$\varepsilon_i = s_{ij}\sigma_j$$

The deformation is expressed as well in the laboratory system by:

$$\varepsilon_{\varphi\psi}^{hkl} = \frac{a_{\varphi\psi}^{hkl} - a_{ref}}{a_{ref}}$$

Where a_{ref} is the free lattice parameter and $a_{\phi\psi}^{hkl}$ is the deformed lattice parameter

III-3. Biaxial stress model

Consider an elastically isotropic material with cubic symmetry. The com-pliances tensor is written as

$$s_{ij} = \begin{pmatrix} s_{11} & s_{12} & s_{12} & 0 & 0 & 0 \\ s_{12} & s_{11} & s_{12} & 0 & 0 & 0 \\ s_{12} & s_{12} & s_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & s_{44} \end{pmatrix}$$

At first, it is assumed that the stress state in the film plane is mainly due to the influence of substrate freezing its lateral dimensions and which gives it a freedom of expansion / compression along the direction of growth. It conseq-uently, the stress terms are zero.

In general, the main constraints $\sigma_{11} = \sigma_{22} = \sigma //$ involve the cancellation of the shear stress and the tensor constraint is written:

$$\sigma_{kl} = \begin{pmatrix} \sigma // & 0 & 0 \\ 0 & \sigma // & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

The evolution of $a_{\phi\psi}^{hkl}$ as a function of $\sin^2\psi$ is linear and is expressed in the case of growth [111] texture by :

$$a_{\psi}^{111} = a_{ref}\left[1 + (2s_{12} + \frac{2}{3}J + \frac{s_{44}}{2}\sin^2\psi)\sigma //\right]$$

Where J is the anisotropy factor expressed as a function of compliances by:

$$J = s_{11} - s_{12} - \frac{s_{44}}{2}$$

And a_{ref} is the reference lattice para-meter without constraints.

A linear regression of experimental points gives us a_{ref} and $\sigma //$

IV. Application To Thin Films Of Y2O3

Sputter deposition is a PVD method of depositing thin films by sputtering. It consist to spry, using an ion beam (Ar in our case) the atoms of the stoichiometic target (Y_2O_3) and deposited on the substrate. For Ion Beam Assisted Deposition, we use the secondary beam for introducing directly other elements (Argon, Oxygen) in the film along its growth. Two electron guns are used for neutralizing the ion beams and avoid their spread. A lamp incorporated into the sample holder allows adjustment of its temperature from ambient to 800 ° C. The chamber is kept under vacuum before the filing (10^{-8} mbar). Spraying is done by an Argon ion beam assisted by $5cm^3$ per minute of oxygen to fix the Y_2O_3 on Si substrate.

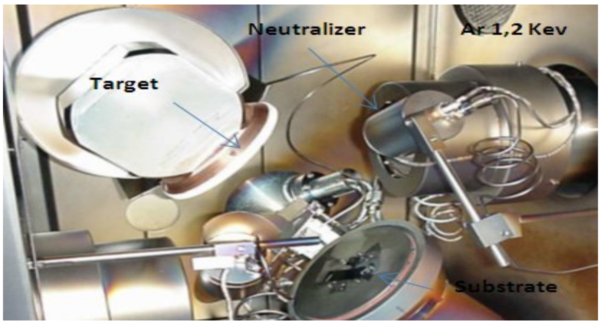

Figure 2: Deposition chamber NORDIKO

Sample 1: Ar at 1200ev assisted by $5cm^3$ of O_2
Sample 2: Ar at 1000ev assisted by $5cm^3$ of O_2
Sample 3: Ar at 800 ev assisted by $5cm^3$ of O_2
Sample 4: Ar At 600 ev assisted by $5cm^3$ of O_2

The analysis of various diffraction peaks of X-ray θ-2θ mode calculates the angle 2θ corresponding to each orientation ψ.

IV-1. Origin of deformation stress states

Hypothesis 1:

The difference in temperature of the substrate and the film creates a bi axial stress

$$\sigma_{th} = \frac{E_{hkl}}{1-\nu_{hkl}}\left(\alpha_s(T) - \alpha_f(T)\right)\left(T_{amb} - T_{dep}\right)$$

E: Young's modulus of the film

ν: Poisson's ratio of film

αf and αs : coefficient of linear expan-sion of the film and substrate

Tam and tdep: Ambient temperature and deposition temperature.

Hypothesis 2:

the film deformation under the action of the substrate (epitaxial relation and difference of lattice parameter) leads to the genesis of the constraint of consis-tency σ_{coh}

Hypothesis 3:

The deposition technique may be the origin of internal stress measured in the layer. The backscattered species play a role in the state of stress in the films deposited by ion beam sputtering. The energy of backscattered particles causes a significant variation of biaxial stress, not the presence of these particles, this phenomenon is due to the Atomic Peening Effect.

IV-2. Kamminga's triaxial model

Step One: the Self focused film is elastically isotropic and its lateral dimensions are equal to those of the substrate. This matrix contains spherical holes evenly distributed and where will be introduced spherical particles of elastic constants identical to those of the matrix.

The difference of rays induces a lattice mismatch (misfit) leading to a change in volume which causes a change in constraints. Thus the deformed lattice parameter is expressed by:

$$a_{def} = a_0\left[1 + \left(s_{11} + 2s_{12}\right)\sigma_{hyd}\right]$$

Step Two: Apply a fixed constraint on the film for equalize its lateral dimensions to those of the substrate, two cases arise

A large effect size

$$\sigma_{hyd} = -\frac{2}{3}\sigma_{fix}$$

A small effect size

$$\sigma_{hyd} = -\frac{(1-\nu)}{(1-2\nu)}\sigma_{fix}$$

Or more generlly :

$$\sigma_{hyd} = -\beta\sigma_{fix}$$

The tensor corresponding to the triaxial stress state is expressed by:

$$\sigma_{kl} = \begin{pmatrix} \sigma_{th} + \sigma_{hyd}\left(1-\frac{1}{\beta}\right) & 0 & 0 \\ 0 & \sigma_{th} + \sigma_{hyd}\left(1-\frac{1}{\beta}\right) & 0 \\ 0 & 0 & \sigma_{hyd} \end{pmatrix}$$ Th

e expression linking the lattice parameter in one direction (ψ, Φ) with different constraints can be written as:

$$a_\psi = a_0\left[1 + \left(s_{11} + 2s_{12}\right)\sigma_{hyd}\right]*$$
$$\left[1 + \left(2s_{12} + \frac{2}{3}J + \frac{s_{44}}{2}\sin^2\psi\right)\left(\sigma_{th} - \frac{\sigma_{hyd}}{\beta}\right)\right]$$

Avec $\beta = \frac{s_{11} + s_{12} - J/3}{s_{11} + 2s_{12}}$

$$s_{11} = \frac{c_{11} + c_{12}}{\left(c_{11} - c_{12}\right)\left(c_{11} + 2c_{12}\right)}$$

$$s_{12} = \frac{-c_{12}}{\left(c_{11} - c_{12}\right)\left(c_{11} + 2c_{12}\right)}$$

$$s_{44} = \frac{1}{c_{44}}$$

$$A = \frac{2c_{44} + c_{12}}{c_{11}} - 1$$

These parameters will be determined by the elastic constants of yttria considered elastically anisotropic because its coefficient of anisotropy A = 0.17 is close to zero.

C_{11}Gpa	C_{12}Gpa	C_{44}Gpa	S_{11} Tpa^{-1}	S_{12} Tpa^{-1}	S_{44} Tpa^{-1}	J (Anisotropy factor) Tpa^{-1}
223.70	112.40	74.60	6.73	-2.25	13.40	2.28

We represent the curve a_ψ as a function of $\sin^2\psi$ and we determine the constraints on each case on using a solver software. The use of software calculations allows to determine the stresses in different cases that arise:

Case 1 :No biaxial stress

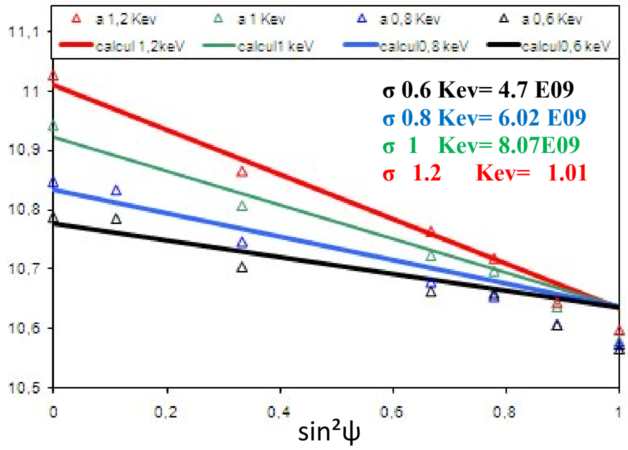

Figure 3 : curve a_ψ for a_0 fixed, β fixed, σth fixed and without σbiax

Case 2 : No thermal or biaxial stress

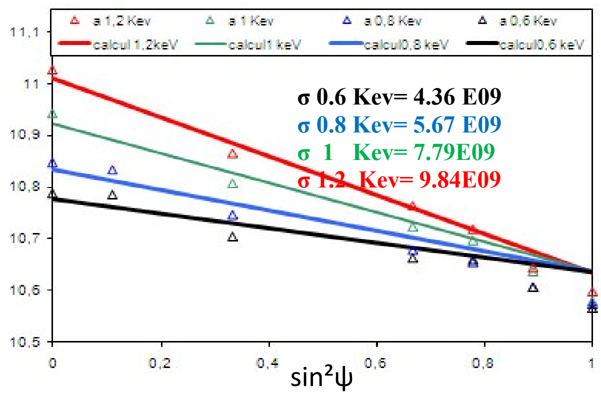

Figure 4 : a_ψ for a_0 fixed, β fixed without σth and σbiax

case 3 : Biaxial constant stress

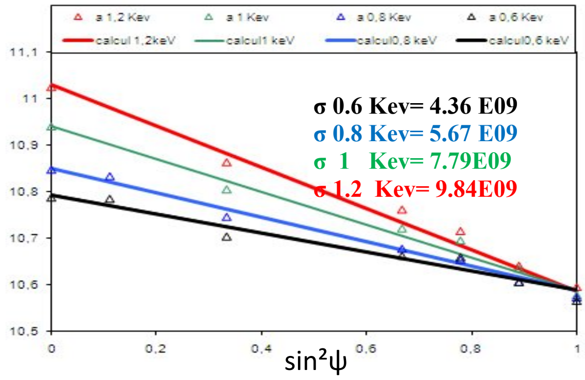

Figure5: a_ψ for a_0 fixed, β fixed, σth fixed and σbiax=-1.2GPa

Case 4 : Biaxial strees variable

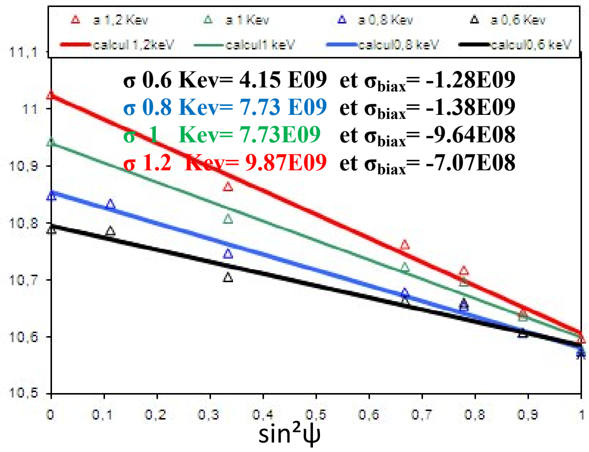

Figure 6 : a_ψ for a0 fixed, β fixed, σth fixed and σbiax variable

Case 5 : Annealing at 300°c and at 700°c

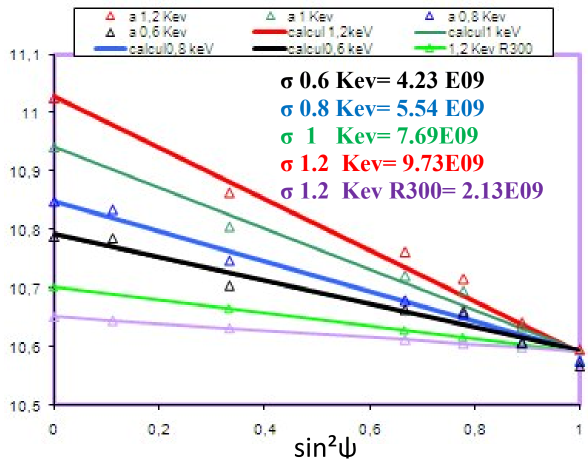

Figure 7 : a_ψ for a0 fixed, β fixed, σth fixed and σbiax=-1.1Gpa

V. Conclusion

The Y_2O_3 thin films deposited on silicon substrate have a high compressive stress associated with free parameters constraints of Fm3m fluorite phase which is the origin of high intrinsic stress; therefore it requires a simulation of high-resolution image to separate the two cubic phases.

Ion Beam Assisted Deposition and the annealing leads in a stress relaxation(- 1.4Gpa) with a low deformation of the mesh.

The electronic diffraction and image simulation on the atomic scale reveal two-phase appearance (Fm3m, Cubic-C) at a very local scale in raw samples highly constrained and appearance phase (cubic-C) unstrained films (annealed films). This is the result of the peening effect, because the bombardment of high energy by the backscattered particles is able to create and stabilize non-equilibrium phases, thus generating a high intrinsic stress.

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