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Critical angles in DC magnetron glad thin films

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ABSTRACT

The objective of this study is to examine the sudden drop in properties of aluminum, titanium and chromium thin films prepared by the glancing angle deposition method. The thin films were deposited by DC magnetron sputtering under identical deposition conditions. A substrate-holder with seven different orientations with respect to the target normal was used. The thickness and the column tilt angles (β) of the thin films were determined by scanning electron microscopy. The residual stress of the thin films was evaluated using the wafer curvature technique and calculated by the Stoney's formula. The thickness variation and column tilt angle versus the orientation of the substrate indicated that the critical point is around 60° for all metallic materials and a critical angle of 60° is also found for the residual stress. Simulations of the particles transport are compared to the experimental data and moderate the critical angles analyses.

1. Introduction

In the recent years, glancing angle deposition (GLAD) technique was employed to control the structure of thin films. By definition, GLAD [1,2] is a physical vapor deposition technique in which the substrate is oriented with respect to the target normal at an angle (α). Also, it undergoes a movement of rotation on itself at an angle φ. According to the variation of these parameters (α, φ), many coatings grow in the form of inclined columns [3,4], zigzag columns [5,6], nano-spirals [7,8], slanted posts [9], nanotubes [10] and branched nano columns [11]. Among the applications of architected films, one can cite optical sensors [12], nano mechanical [13], pressure sensors [14], and field emitters [15]. Most of the researches concerning GLAD layers were applied to pure material such as Si [7], Cr [4,16], Pt [17], Ta [11,18], W [7], Ti [19] and Co [7]. Others preferred to work on oxides such as TiO_2 [20,21], SiO_2 [21], ZrO_2 [22], or sulfides such as ZnS [23] and Sb_2S_3 [24].

Most of the properties present a critical point (or zone) around 50° where a change is observed [25–27]. In these papers, the results are often compared to the atom incidence angle, which is assumed to be equal to the substrate orientation angle and the critical zone is only noticed as a particular characteristic. Due to the numerous deposition conditions, chamber and target geometry, material deposited, etc., a global understanding of the mechanisms ruling the critical zone is limited.

The aim of this paper is to highlight the presence of critical zones in some properties of inclined columnar coatings. To reduce the number of variables in the system, different substrate inclinations are used during one deposition under constant conditions (temperature, pressure, discharge power, etc.) for three pure metallic coatings. For each property the critical angle is discussed. Simulations are computed and give the number of particles detected for each angle, the angle of incidence and the energy of the impinging particles.

2. Experimental

Aluminum, chromium and titanium thin films were deposited on (100) Si substrates (20 mm × 10 mm × 0.38 mm) with a DC magnetron sputtering system (KENOSISTEC - KS40V). The working pressure was set at 0.09 Pa ± 0.005 to limit the scattering during gas transport. Indeed, in these conditions the mean free path is about 74 mm and with a target-to-substrate distance of 105 mm, the average number of collisions is 1.4. Rectangular targets (407 mm × 127 mm × 6.35 mm) of the three metals are used: aluminum (purity of 99.99%), chromium (purity of 99.95%) and titanium (purity of 99.99%). The negative discharge voltage of the aluminum, chromium and titanium targets was 464, 402 and 636 V

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respectively with a constant power of 1500 W. The substrate-holder carries seven orientations $\alpha = 0°, 15°, 30°, 45°, 60°, 75°$ and $85°$ for each deposition with respect to the target normal (Fig. 1).

α is the angle between the substrate and target's normal. Positions of the substrate on the holder are chosen in order to minimize self-shadowing. Each holder has a width of 2 cm and the distance between two holders is 1 cm. The deposition time was 16, 19 and 35 min for aluminum, chromium and titanium coatings respectively. These durations were chosen in order to obtain 1 μm thick layers for samples with an angle $\alpha = 85°$. This minimal value ensures that the stress measurements will be less influenced by the thickness variations [28].

The morphology of the cross section and the surface of the thin films were analyzed with a scanning electron microscope (SEM-Jeol JSM-5900 LV). From these observations, the thickness and the column angles (β) with respect to the substrate normal in columnar microstructures of the thin films were determined. One should notice that increasing the substrate inclination leads to a gradient in the thickness over one direction, i. e over the direction target-substrate in the plane of both target and substrate normal (cut plane of Fig. 1). Indeed, smaller the target/substrate distance is, thicker the films are [16]. The film thickness mentioned in this study is an average value. The overall maximum variation of the thickness measurements is ±0.1 μm.

The residual stresses of the thin films were evaluated using the wafer curvature technique and the modified Stoney's formula for a plate like substrate [29] was applied (Equation (1)):

$$\sigma = \pm \frac{E}{6\,(1 - \upsilon)} \times \frac{t_s^2}{t_f}\left(\frac{1}{R}\right) \qquad (1)$$

where E is the substrate's Young modulus, υ is the substrate's Poisson ratio, t_s is the substrate's thickness and t_f is the thickness of the thin films producing the stress, R the radius of curvature of the bended surface. The substrate curvatures, i.e. the topographic images of the whole substrate [30], before and after deposition were measured by optical profilometry (VEECO, Wyko NT-1100). The subtraction of the initial curvature image to the coated one allows obtaining the topographic image of the substrate deformation from where the radii are then extracted. This process and the extraction of the radii of the principal directions are performed by the Gwyddion software [31].

The incidence angle of the incoming particles was computed by SIMTRA [32]. The calculations are based on the experimental settings (pressure, geometry and target erosion profiles) and TRIM calculations [33]. The angle used in this paper is calculated from the resultant vector of the particles impinging the substrate. Thus it is representative of the in-plane and the out of the plane distribution and also of the position and orientation of each substrate compared to the center of the target.

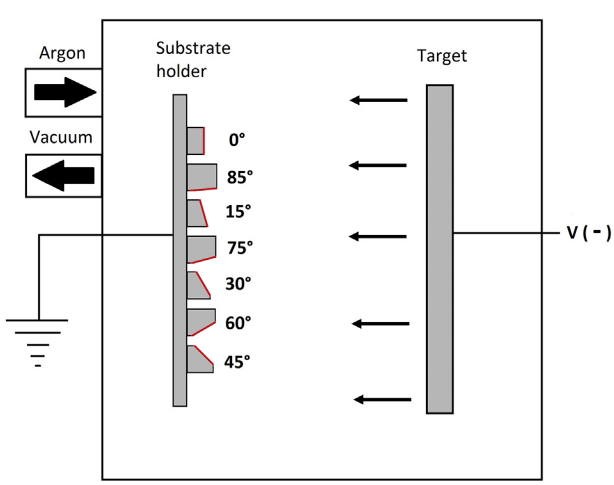

Fig. 1. Chamber description: On the left the substrate-holder is presented in the experimental configuration. The target and the substrate-holder are centered.

3. Results and discussion

3.1. Thickness

The normalized thickness of the Al, Cr and Ti thin films and the normalized number of particles detected from SIMTRA calculations varying with the substrate inclination angle are illustrated in Fig. 2. When the substrate is facing the target, the thickness of the Al, Cr and Ti coating is about 2.5, 2.8 and 2.35 μm respectively.

The thickness of each coating depends on the number of atoms captured by the substrate surface. This amount is linked to the projected surface (parallel to the target plane), which evolution according to the orientation of the substrate is ruled by a cosine law, cos (α), and will serve as a reference.

Two behaviors could be observed for the experimental data (Fig. 2-a): for $\alpha \le 60°$, the thickness of the Al, Cr and Ti films decreases while the substrate inclination angle increases. The thickness evolution of the three materials follows globally the cosine law. Aluminum is the closest, where chromium is slightly higher. Titanium begins with a clear increase and then decreases with a higher slope than the cosine law. For these variations, a law, $\cos^s(\alpha)$, proposed by Woo et al. [20] could fit the series. *s* is a coefficient that takes into account the materials and the deposition conditions, without any direct physical signification. For $\alpha \ge 60°$, the thickness of the films remains almost constant. The number of particles computed from SIMTRA calculations shows a similar trend with substrate inclination as the film thickness variation (Fig. 2-b). Below an angle of 60°, a cosine-like evolution is observed, although the values are higher than the cosine law. For $\alpha \ge 60°$ computed values level off, and this effect is more pronounced for titanium than for chromium and aluminum. One can notice that the calculations predict more particles for $\alpha = 15°$ whatever the material. This is measured experimentally for chromium and titanium films. Especially for titanium, an increase of 10% compared to the normal deposition is reported.

Deviation from the cosine law has been also reported but mostly with a more continuous profile than the one reported here and often explained by a variation of the film mass density and a material effect [23,24,34–36]. In our case, the target could not be considered as a perfect punctual source as supposed in the cosine law. Consequently, the particular transport conditions, i.e. the scattering during transport, the shape of the racetracks, the size of the targets, the position of the samples relative to the center of the targets and a self-shadowing effect induced by the substrate-holders, will moderate the simple cosine law that only depends on the substrate inclination angle. Fig. 3 present the calculated normalized number of particles versus the experimental normalized thickness.

A good linear correlation between the experimental and calculated results is found for all materials. Taking into account all the process parameters mentioned above and according to our particular system configuration, SIMTRA calculations confirm the influence of the particles transport on the thickness of the films. Some dispersion is observed between the thickness and the calculated number of particles and the particular behavior of each material. But, it is noteworthy that the thickness of a film is not only due to the number of impinging particles but also to the film morphology.