Model of texture development in polycrystalline films growing on amorphous substrates with different topographies

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Abstract

A two-dimensional computer model is presented to study the development of microstructure and crystallographic texture in polycrystalline films growing on amorphous substrates, which often exhibit strong textures. In particular, the analysis focuses on the influence of growth parameters such as the crystal habit, the relative growth rate along different crystal directions, the nucleation density on the substrate surface and the substrate topography. The simulation explains the development of fiber and in-plane textures in polycrystalline films growing from randomly oriented nuclei. These textures result from a geometric selection of the orientation of crystals, mainly due to differences in growth rates along different crystal directions. The calculated distribution of crystal orientations and its evolution with time are in agreement with textures observed in films deposited under growth conditions, corresponding to zone 2 of the structure zone model. The growth of crystals composing of a polycrystalline film is mainly constrained by the growth of adjacent crystals and the geometry of the environment where crystal growth takes place. For instance, average lateral crystal size scales with time as a power law, the exponent varying as a function of the curvature and/or topography of the substrate. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Polycrystalline thin films usually exhibit strong crystallographic texture (non-random or preferential orientation of crystals). It is also well known that the properties of polycrystalline materials depend strongly on their microstructure (grain morphology and size) and on crystal orientation (crystallographic-texture) [1–4]. For instance, it has been observed that the lifetime of thin film conductors depends on both the mean grain size and texture [5,6]. Therefore, controlling the microstructure and texture is critical when fabricating polycrystalline thin film devices.

The preferential orientation of crystals or texture may result due to a directional process such as the heat flow during directional solidification, plastic flow during deformation (i.e. lamination, extrusion, tape casting) or atomic flux during film deposition [1,4]. In other cases, as during recrystallization and grain growth, texture evolves as the result of the preferential growth of some grains (abnormal grain growth) having a certain crystallographic orientation [7]. On the other hand, during texture formation, the role of the substrate can be active (e.g. epitaxially induced two-dimensional nucleation) or passive (e.g. crystals growing on an amorphous substrate). In the first case, the orientation of the film is the same as the substrate (parallel growth).
In the second case, the preferential orientation may result from a geometrical selection of the orientation of crystals during growth, due to differences in the growth rates along the different directions in a crystal.

As described by Grigoriev for mineral aggregates (i.e. quartz geodes) [8] and van der Drift for thin-films [9], during growth, crystals compete for the available space and only those favorably oriented, that is with their faster growth direction perpendicular to the substrate, are selected and contribute to the outer surface. Finally, all crystals will have aligned this crystal direction but will be rotated in azimuth (fiber texture). More generally, crystals will orient those with their fastest growth directions parallel to the direction of advancement of the growth front. This mechanism named ‘evolutionary selection’ or ‘competitive growth’ explains the preferential orientation of crystals starting from randomly oriented nuclei. Our simulation is based on this mechanism, which is the dominant one during the microstructure development in films deposited at intermediate temperature ($0.3 < T_s/T_m < 0.6$; zone 2 of the structure zone model) [10,11]. $T_s$ is the substrate temperature and $T_m$ is the melting point of the film material. Under these conditions, there is not enough bulk diffusion to restructure the film microstructure once it forms, and a purely geometrical model, as the one described here, can be used. In contrast, at the lower part of the zone 2, amorphous or nanocrystalline films are formed with weak or no preferential orientation of crystals, unless channeling or resputtering processes occur [12–14]. In this lower region, the governing processes are ballistic aggregation, limited surface diffusion, and self-shadowing [15]. At a higher temperature ($T_s/T_m \sim 0.5–0.6$) region, where surface diffusion is high and self-shadowing effects are minimized, large grain size, oriented columnar crystallites are formed. For even higher temperatures ($T_s/T_m > 0.6$, zone 3) bulk diffusion starts and increases with temperature, and the dominant processes are grain growth and boundary migration [16,17].

In this paper, we use a computer model based on the competitive growth mechanism to study the evolution of the microstructure and texture in polycrystalline films. Previously, Thijssen et al. [18,19] and Wild et al. [20–22] used a similar model to study the evolution of diamond films. Their models basically consider simple square crystals growing on flat surfaces. In contrast, the model presented here also explores the effect of several important parameters such as different crystal habits and symmetries, the anisotropic growth along different crystal directions and the substrate topography. It also calculates the distribution of crystal orientations and how is it affected by the previously mentioned parameters.

2. Description of the model

In our model [23,24], every crystal is represented as a polygon and every face of the crystal as a segment of a line, $L$, delimited by two vertices, $v1$ and $v2$, which are the intersecting points of neighboring facets. The line is defined by the parametric equation, $Ax + By + C = 0$. Crystal growth consists of parallel displacement of faces at constant growth increments (see Fig. 1a). After each growth increment, the crystal faces are defined by a new line parallel to the previous face and displaced a distance, $d$. Crystal morphology is determined by the relative rates of growth of the crystal along different

![Fig. 1. Growth of a polycrystalline film: (a) section of a growing single crystal consisting of prismatic (10), pendion (01) and pyramidal (11) forms. The ratio of the growth rates of the different faces are: $\alpha = G_{[11]} / G_{[10]} = 0.9$ and $\gamma = G_{[01]} / G_{[10]} = 2.0$. The crystals are elongated along the [01] direction, which is the faster growth direction; (b) sequence of the nucleation and texture formation in a polycrystalline film.](image-url)
directions. For crystals composed of \{01\}, \{11\} and \{10\} faces, we can define the ratios of the growth rates of these faces as:

\[
\alpha = \frac{G_{[11]}}{G_{[10]}}
\]

\[
\gamma = \frac{G_{[01]}}{G_{[10]}}
\]

where \(G_{[hk]}\) is the rate of growth of \([hk]\) faces. For instance, as the value of \(\gamma\) increases, the crystals become more elongated along the \[01\] direction. The orientation of a crystal is defined by the tilt angle, \(\chi\), of the \[01\] direction with respect to the substrate normal (see Fig. 1a). In our model, initially, crystal seeds are set on a line, which acts as a substrate, with a given orientation (i.e. random) and intersected spacing, \(\Delta\). As the crystal seeds grow, adjacent crystals contact one another, intergrain boundaries (along the bisector of the two meeting faces) form and a continuous film results. As film growth proceeds, some crystals overgrow others so that the film surface is constantly changing.

At each growth increment, the program computes the outer surface of the film, as the envelope of the crystals growing in contact (Fig. 1b). To do so, the program computes the intersection points of each face with its adjacent one, as the intersection point of the two lines defining the two meeting faces. This process is repeated as the program scans all film surface, from the first crystal (left) to the last one (right), similarly as one would draw it by hand. It also counts the number of crystalline domains, or crystal grains forming the film surface, measures their size and orientation (crystallographic texture). The main growth parameters that can be varied are: (a) the starting crystal morphology or habit; (b) the orientation of crystals; (c) the relative growth rate of the different crystal faces; (d) the nucleation density on the substrate, and (e) the substrate topography. Thickness is normalized in units of the average distance, \(\Delta\), between two neighboring nuclei. In our model, the deposition rate is constant and we can study the evolution of the film either as a function of its thickness or time of growth.

Texture is determined experimentally by measuring a given \(hkl\) Bragg reflection at different sample orientations, using an X-ray pole figure diffractometer. For a given sample orientation, the recorded intensity of the selected \(hkl\) reflection is proportional to the number of crystals, having their \(hkl\) lattice planes oriented in the condition of Bragg reflection [25]. Due to X-ray absorption, the main contribution to the intensity arises from the outer surface of the aggregate. Similarly, our program computes the orientation distribution of the \(hk\) planes of crystals, composing the film surface. To do this, we measure the length of the \(hk\) planes, \(S(\chi)\), that are oriented within a given interval \(\{\chi - \Delta\chi, \chi + \Delta\chi\}\), where \(\chi\) is the tilt angle of the normal to the plane with respect to the substrate normal. Texture information is represented using rocking curves (two-dimensional section of a pole figure), displaying the statistical orientation of \(hkl\) lattice planes relative to the substrate surface. The scattering or degree of preferential orientation of the crystals can be measured as the full-width at half-maximum (FWHM) of the peaks of the \(\chi\) profile of the measured distribution. The lower the values of FWHM, the higher the alignment of crystals.

3. Results and discussion

Fig. 1b illustrates the formation of a polycrystalline film through a sequence of growth steps. In this case, the polycrystalline film is growing from randomly oriented seeds settled on a flat substrate. Initially, crystal seeds grow freely. Later, adjacent crystals meet and form intercrystalline impingement boundaries. During subsequent growth, only those crystals oriented with faster growth directions (in this case, \[01\] direction) perpendicular to the substrate surface grow and contribute to the outer surface of the film. Crystals with other orientations are buried by neighboring crystals with a more favorable orientation. This process results in a decrease in the number of crystalline domains that are not preferentially orientated and an increase in their size (Fig. 2). During growth, three different stages of microstructure development can be differentiated, namely: (1) isolated growth, which is characterized by the free growth of the starting nuclei; (2) competitive growth, when the crystals compete for space and selection of the best crystal orientation occurs; and (3) parallel growth, the final stage when only (or mainly) those crystals with proper orientation continue to grow. The resulting film morphology is columnar with blade-shaped single crystal domains, similar to that of a cross-section of hexagonal AlN films [26,27].

3.1. Development of out-of-plane or fiber texture

Fig. 3 shows the evolution of the orientation of \(01\) faces (or 01 rocking curve) for a film composed of elongated crystals (see Fig. 1a). Initially, the crystallites are randomly oriented (being \(S(\chi)\) constant), but as the film grows thicker, one maximum arises at an angle of \(0^\circ\), corresponding to crystal units oriented with \(01\) faces parallel to the substrate surface. Therefore, the crystals contributing to the growth surface are those with \(01\) directions oriented perpendicular to the growth front of the film, resulting in a strong \(01\) fiber texture. As film thickness increases, the peak height increases and FWHM decrease sharply, initially, and later more slowly, indicating an increasingly preferen-
Fig. 2. Evolution of the microstructure of a film composed by crystals with different habits: (a) the number of crystalline domains which forms the outer surface of the films; and (b) their average lateral size.

3.2. Development of a sheet or in-plane texture

Crystals nucleating on an amorphous substrate do not have to be randomly oriented but can nucleate with an orientation which minimizes surface and interface energy [28]. This mechanism may account for the development of a fiber texture (i.e. with a [111] and [001] orientation for cubic and hexagonal systems, respectively) where crystals nucleate on the most compact crystal planes. However, to achieve an in-plane orientation, where crystals have the other two crystallographic axis aligned in the plane of the growth, another complementary mechanism has to be invoked (i.e. competitive growth). For instance, during a directional solidification [29,30], an in-plane orientation may be obtained if crystals nucleate with their minimum energy faces parallel to the substrate; and additionally, the growth (or solidification) front advances parallel to the substrate. In this case, due to a competitive growth, crystals will orient with their faster growth direction parallel to the direction of the advancing growth front, which is parallel to the substrate surface (see Fig. 4). Then, a \([h_1k_1l_1]\) \((h_mk_ml_m)\) sheet texture or in-plane orientation will result, where \(h_mk_ml_m\) corresponds to the indices of the crystal face with the lowest surface energy and \(h_1k_1l_1\) to those of the direction of faster growth, which will be oriented parallel to the growth direction. Similarly, Karpenko et al. [31] explain the development of an in-plane texture of off-normal sputter deposited thin films by a cooperative effect of self-shadowing and the anisotropy of crystal growth rates. Other mechanisms proposed are based on channeling effect and resputtering of specific crystal faces [13,32,33].

3.3. Influence of the crystal habit and anisotropy of the crystal growth rates

Since the selection of crystals is purely geometric, the habit of crystals plays an important role in the development of microstructure [8,34]. The habit of the crystals is determined by the relative growth rates of the different crystal directions, which are controlled by the deposition conditions (growth temperature, absorption of impurities, surface energy and diffusion) [26,35,36]. Only the slow growing faces contribute to the growth morphology of crystals. However, the growth of a polycrystal is much more complicated. The selection of the orientation of the crystals forming the film surface is due to differences in the growth rates along different crystal directions in each crystal, i.e. the anisotropy of the growth rates. As stated above, the preferential orientation of films coincides with the fastest growth direction. Furthermore, as the anisotropy increases (from rounded to elongated crystals), the rate of decrease in the number of crystals, forming the film surface increases as well as the rate of texture development.

For rounded crystals (nodular growth), there is no selection during their growth. For any other crystal shape (e.g. square, rectangle, rhombohedral, prismatic, etc.) selection occurs and the three stages described above appear consecutively. In Fig. 2a, it is apparent that for crystal shapes other than circles, the number of crystals starts to decrease sharply and subsequently the rate of selection slows down. The number, \(N\), of surviving crystals decreases while their lateral size, \(S\), in-
creases with time or thickness, $t$, according to the power law:

$$N = N_0 (t + 1)^{-\beta}$$

(3)

$$S = S_0 (t + 1)^{\beta}$$

(4)

where the exponent $\beta$ takes a value between 0.58 and 0.50, in agreement with previous results [18,19]. However, in the case of a film composed by rounded crystals (nodular growth), the number of crystalline domains and their sizes remain constant during growth (see Fig. 2).

Fig. 5 shows the dependence of film orientation on the relative growth rates of different crystal faces. In Fig. 5a, as the value of $\gamma$ increases from 1.0 to 2.0, the orientation of the aggregate changes from a [11] fiber texture to one along the [01] direction. In the pole Fig. 5a1, two maxima arise at inclination angles of $\pm 45^\circ$, corresponding to crystal units oriented with their (01) planes tilted $45^\circ$. Note that the angle separation between [11] and [01] directions is $45^\circ$. Therefore, crystals are oriented with [11] directions perpendicular to the substrate surface. Fig. 5b illustrates schematically how film texture varies depending on the relative growth rate of the different faces. Three regions with different crystallographic orientation can be differentiated. For values of $\gamma = 1$ and $\alpha < \gamma$, the [01] direction is the fastest growth direction and the film orients after (01). For values of $\alpha > 1$ and $\gamma > \gamma$, the [11] direction is the fastest and the film orients after (11). Finally, for values of $\gamma = 1$ and $\gamma < 1$, the [10] direction is the fastest and an orientation after (10) results. The degree of orientation increases in the direction of the arrows.

### 3.4. Influence of substrate topography

The available crystal growth space, forming part of a film, is limited by the growth of adjacent crystals and also by the geometry of the environment where growth takes place. Crystals growing on a convex substrate have an increasing available space to develop while those growing on a concave substrate, or cavity (i.e. trench), have less accessible space and an increasing competition for it. Therefore, depending on the topography of the substrate, the microstructure and texture evolution will be different [37].
Fig. 5. (a) 01 pole figures (rocking curves) of films composed of crystals with different habits and relative growth rates of: (a1) $\alpha = 1.3$, and $\gamma = 1.0$; (a2) $\alpha = 1.0$, and $\gamma = 1.0$; (a3) $\alpha = 0.89$, and $\gamma = 2.0$. (b) Schematic showing the dominant texture for different relative growth rates.

Fig. 6 shows the growth pattern of films grown on different substrate topographies. On a trenched substrate, crystals grow homogeneously all over the substrate, filling the trenches. The height advantage and extra available space for crystals growing on the top features allow them to prevail and overgrow those growing inside of the trench, developing a fan-like morphology (see Fig. 6b). On curved substrates, for a concave substrate or cavity, the morphology is geode-like while for a convex one it is druse-like (see Fig. 6c). In all cases the microstructure is columnar.

When studying the variation of lateral size of crystalline domains (column width) with time, there are notable differences depending on substrate topography. Fig. 6d shows that the size of crystals growing inside a trench increases until reaching a maximum size, when crystals from opposite walls meet and stop growing. On the other hand, the size of crystals growing on the top of the trench steadily increases, their growth rate being higher than those growing inside the trench. For crystals nucleating on a convex or plane substrate, average crystal size scales with time as a power law, of exponent $\beta$, varying from 0.65 for a convex to 0.5 for a plane substrate. The size of crystals growing on a concave substrate or cavity initially increases, then reaches a plateau at a maximum value and finally decreases. In this final stage, crystals collapse and growth ends, as the whole space of cavity is filled (see Fig. 6e). The above dependence of crystal grain size on substrate curvature has been described by Trava-Airoldi et al. [38], for diamond thin films growing on substrates of different shapes.

Also in directional solidification, the shape of the growth front affects the crystal perfection of a film. In laser-beam crystallization, crystals grow perpendicular to the liquid–solid interface (growth front) [30]. Then, if the laser produces a concave growth front, smaller crystals will form the resulting film. On the other hand, if the growth front is convex, wider crystal domains will form and a film of better quality will result.

4. Conclusions

Although, the model is simplistic, it does reflect the anisotropic nature of thin film growth. It satisfactorily explains the development of microstructure and texture, in polycrystalline films, due to a process of geometric selection of crystals according to their orientation. The calculated angular distribution of crystal orientations and its evolution over time are similar to those observed experimentally for polycrystalline films. The model could be useful in interpreting experimental distribution of crystal orientation and to estimate some growth parameters such as relative growth rates, which in turn will help in understanding the deposition conditions.

In contrast to earlier models, the model presented here additionally explores the effect of other parameters such as different crystal habits and symmetries, the anisotropic growth along different crystal directions and the substrate topography. Anisotropy of the crystal growth rates, the arrangement of neighboring crystals (i.e. nucleation density) and substrate topography mainly influence the evolution of the microstructure of the films. The selection of the orientation of the crystals is due to the difference in the growth rates along different directions in a crystal. The preferential orientation of a film coincides with the fastest growth direction. Furthermore, as the anisotropy increases (from rounded to elongated crystals), the rate of decrement of the number of crystals forming the film surface increases as well as the rate of texture development. Since preferential orientation of the film is determined by the relative growth rates of crystal directions, it can
be altered by selective absorption of impurities that will retard growth along specific crystal directions.

The growth of crystals composing a polycrystalline film is limited by the available space, which is mainly constrained by the growth of adjacent crystals and the geometry of the environment where growth takes place. The substrate topography strongly influences the microstructure development of polycrystalline films as it constrains the free space available for the growing crystals. Average lateral crystal size scales with time as a power law, the exponent varying as a function of the curvature and/or topography of the substrate.

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References


Fig. 6. Microstructures of polycrystalline films grown on substrates with different topographies: (a) a flat substrate; (b) a trenched substrate; and (c) a concave substrate. Evolution of the lateral size of the crystalline domains (columnar width) as a function of film thickness for a film growing: (d) on a trenched substrate; and (c) on a convex, flat or concave substrate.


