Thin Solid Films 604 (2016) 90–93

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Critical review

Quantitative correlation between intrinsic stress and microstructure of thin films

D. Depla, B.R. Braeckman

Department of Solid State Sciences, Ghent University, Krijgslaan 281/S1, 9000 Ghent, Belgium

ARTICLE INFO

Available online 23 March 2016

ABSTRACT

A critical review of the available literature on thin film intrinsic stress has generated a database with 111 entries representing 19 different metals deposited by evaporation. Although there is a wide range of experimental conditions, the data can be presented in a comprehensible way based on the surface mean free path of diffusing adatoms. This characteristic length L is calculated based on the deposition temperature, the melting temperature of the evaporant, and the deposition flux. The calculated strain as a function of L not only shows the trends in a quantitative way, but also allows one to connect the data with the thin-film microstructure as represented in structure-zone models. The proposed procedure appears to also be applicable to amorphous metallic glass thin films (4 alloy systems, 29 entries) as well.

Contents

1. Introduction

Intrinsic stress evolution during thin film deposition by evaporation is a research topic that has been studied for decades and has been reviewed in several papers [1–9]. The intrinsic stress is strongly material-dependent, and for continuous, non-epitaxial thin films can be divided into two groups. For materials deposited under conditions leading to low adatom surface mobility, a tensile stress state is observed, while a compressive stress state is usually obtained for high-adatom-mobility materials. For metals deposited at the same substrate temperature T_s, the evaporant melting temperature T_m is a good measure to distinguish between these two classes since diffusion processes for metals scale inversely with T_m. The Arrhenius behavior of these processes explains the importance of the deposition (or substrate) temperature T_s, and hence experimental trends are often presented in terms of the homologous temperature, i.e. the ratio between the deposition temperature and the melting temperature, both expressed in Kelvin (T_s/T_m). The homologous temperature is also used to classify the microstructure of evaporated thin films in structure-zone models (SZM). A relationship between the microstructure and the intrinsic stress of thin films can therefore be expected. Despite this apparent connection, no quantitative proof has been given in the literature. Here this quantitative link is made and this allows one to predict the stress state of evaporated thin films.

2. Method

Kinetic models used to describe thin-film nucleation [11] predict scaling laws for the mean free path of the diffusing adatoms before they form a nucleus or are captured by an existing island, also known as the characteristic nucleation length L. When the critical nucleus size is one atom, which is a quite common situation during thermal evaporation, L is related to the incident flux of atoms F [11–13]

$$L = \sqrt{\frac{1}{\eta}}\left(\frac{Da_s^2}{F}\right)^{1/6} \quad (1)$$

with D the adatom surface diffusion rate, a_s the lattice parameter of the surface unit cell, and η a dimensionless parameter with a maximum value of 0.25.

http://dx.doi.org/10.1016/j.tsf.2016.03.039
0040-6090/

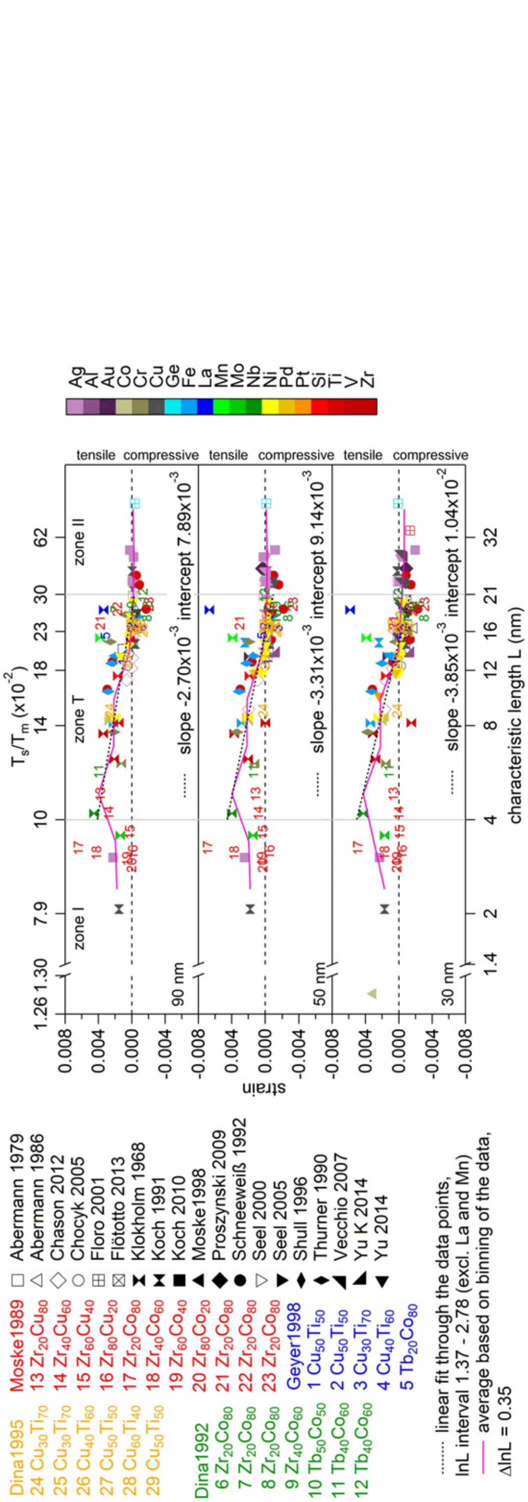

Fig. 1. Strain, measured at different film thicknesses, as a function of the characteristic length L (lower horizontal axis). The symbols refer to the references while the color code is used to identify the material. The upper horizontal axis converts the characteristic length L to the homologous temperature via Eqs. (1) and (2) in the text, using a deposition flux F averaged over all references. To calculate the average strain (solid line), the data was ordered according to the characteristic length L with a bin size of $\Delta \ln L = 0.35$. The maximum and minimum of the average strain correspond approximately to a homologous temperature of 0.1 and 0.3 respectively, which are also the demarcation temperatures between zone I and zone T, and zone T and zone II, in the structure zone model of Sanders [10]. A linear correlation was obtained between the strain and the natural logarithm of the characteristic length for thin films in a tensile stressed state within zone T. This correlation is indicated with a dashed line. The symbols refer to pure metals while numbers are used to identify amorphous metallic glasses.

The surface diffusion rate depends on the homologous temperature, and can be written as

$$D = D_0 e^{-CT_m/T_s} \tag{2}$$

according to Flynn [14]. D_0 and C are 2×10^{-4} cm^2 s^{-1} and 1.5 respectively for closest-packed metal surfaces. With these equations, it is possible to evaluate the impact of the deposition flux on the characteristic length. A decrease in the deposition rate by a factor of 10, results at $T_s/T_m = 0.2$, in an increase in L by approximately 50%. This increase in L can also be achieved by increasing the homologous temperature from 0.200 to 0.288, or stated differently with a substrate temperature increase of 44%. Some papers on intrinsic stress during thin-film deposition explore the material dependency without considering the difference in F, although it plays a pivotal role in thin-film microstructural evolution. This could be one of the reasons for the missing quantitative relationship between microstructure and intrinsic stress.

Eqs. (1) and (2) enable one to compare experiments performed with different metals, at several deposition temperatures, and with different deposition rates. The following procedure was used. Intrinsic stress measurements are in most cases presented as graphs of force per unit width (N/m) vs. film thickness (nm) [8,9,15–31]. Here, the graphs have been digitalized, and the first derivative, or the film stress, was determined at three different film thicknesses (30, 50 and 90 nm), where available, by a linear fit through five data points. From the obtained stress values, the strain was calculated using the biaxial modulus $(E / (1 - \nu)$, with E the Young's modulus and ν Poisson's ratio. As Eq. (2) is only valid for closest-packed surfaces, it was assumed that the films have a fiber texture with the closest-packed lattice planes ((111) for fcc metals, (110) for bcc metals and (0001) for hcp metals) parallel to the substrate surface [32–34]. As the closest-packed surfaces typically have the lowest surface energy [35], this assumption seems reasonable. The anisotropy of the biaxial modulus [34,36] was taken into account to calculate the strain. The surface lattice parameter was calculated from the metal's unit cell parameters, and used together with the deposition flux and the diffusion rate to obtain the characteristic length L from Eq. (1).

3. Results and discussion

Fig. 1 presents literature film strain results as a function of the characteristic length L (note that L is plotted on a log scale) for the three film thicknesses noted above. To show the overall trend, the data was binned according to the natural logarithm of the characteristic length with a bin size of $\Delta \ln L = 0.35$, and the average strain was calculated per bin. The results are plotted as a solid line in Fig. 1. A slight increase in the tensile stress is observed at low L values, reaching a maximum at L ≈ 5 nm. At higher values of L, the tensile stress decreases to become zero at L ≈ 14 nm. Films with L ≥ 14 nm are in compressive stress reaching a minimum (for 30-nm- and 50-nm-thick films) at approximately 28 nm. Above this latter value, the films again become somewhat more tensile, or at least less compressive.

To link this trend with the microstructure, the SZM of Sanders [10], which is a refinement of the initial SZM of Movchan and Demchishin [37], has been used. Sanders defines three important zones. Zone I is the low-adatom-mobility zone, often called the hit-and-stick regime, which is characterized by a porous microstructure. The transition zone T is defined as T_s/T_m between 0.1 and 0.3. The out-of-plane growth competition between grains with different crystallographic orientations results in a V-shaped columnar microstructure. At $T_s/T_m > 0.3$, grain boundaries become mobile [38], and the microstructure changes to a thin film composed of domed, straight columns with larger width. An overview of this, and other, SZMs is given by Mahieu et al. [38].