11th International Conference on Advanced Materials
ICAM 2009
Rio de Janeiro Brazil
September 20 - 25

Growth of nanocomposite films: from dynamic roughening to dynamic smoothing

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Abstract – This paper reports several new findings on the breakdown of dynamic roughening in thin film growth. With increasing energy flux of concurrent ion impingement during pulsed DC sputtering, a transition from dynamic roughening to dynamic smoothing is observed in the growth behavior of TiC/a-C nanocomposite films. The nanocomposite films show a negative growth exponent and ultra-smoothness (RMS roughness ~0.2 nm at film thickness of 1.5 μm). We were able to predict the evolution of surface roughness based on a linear equation of surface growth which contains two diffusivity parameters that control the atomic mobility along the growing outer surface. The model is in good agreement with atomic force microscopy measurements of roughness evolution.

Dynamic roughening is commonly observed in plasma processing of materials, including both film deposition and plasma etching. In fact the roughness of a growing outer surface increases with processing time and film thickness. This so-called dynamic roughening can be attributed to the fluctuation of incoming particles (noise) and shadowing effects. As a result a rough surface will lead to columnar growth such that the column boundaries (CBs) originate at the networks of grooves on the growing surface and the hills become the spearheads of the columns. Such a columnar structure is undesired because the CBs are the potential source of failure, e.g. mechanical (cracking), chemical (corrosion) and thermal (oxidation). In contrast, ultra-smooth surfaces produce pinhole-free films which are of considerable technological importance, e.g. in magnetic disk storage systems.

In this paper it is shown that with increasing intensity of concurrent ion impingement by raising the frequency of pulsed DC sputtering, a transition from dynamic roughening to dynamic smoothing is revealed in the growth behavior of TiC/a-C nanocomposite films. The dynamic smoothing of growing surface in thick TiC/a-C nanocomposite films is characterized by a negative growth exponent and ultrasmoothness. As a solid proof of the impact-induced downhill flow model and subplantation model, an amorphous front layer of 2 nm thickness is observed to always cover the bulk nanocomposite. The formation of such an amorphous front layer excludes any influence of nanocrystallites on the dynamic growth behavior and roughness evolution of the nanocomposite films. The predicted evolution of surface roughness based on a linear equation of surface growth which contains two gradient terms is in good agreement with the atomic force microscopy measurements of roughness evolution.

Combined XTEM and nanoindentation investigations were carried out as well and the following conclusions can be drawn:

- The nanocomposites show excellent toughness while maintaining high hardness. By using proper deposition parameters potential damage along shear bands and a-C sublayers can be avoided. Nanocracks are observed in the nanocomposite with the highest Ti content, but are found to be restricted only to the surface region and are halted by the nanograins.
- Multiple shear bands uniformly nucleating within the nanocomposite and propagating over a small distance were observed in the top layer. Shear deformation was brought under control by the nanocompositing structure, with each band contributing to plastic deformation but conveying a shear strain not high enough to cause damage.
- In addition to the uniformly nucleated multiple shear banding, shear delocalization was observed to occur in the nanocomposite toplayer. The abrupt shear strain arising from the interlayer was accommodated in the nanocomposite top layer through a delocalizing process, which spreads the shear bands to an increasing width and thereby suppressing crack propagation.