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Effects of oblique-angle deposition on intrinsic stress evolution during polycrystalline film growth

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Abstract

Precise control of the residual stress in polycrystalline films, which remains a central but difficult task in a variety of emerging technologies, requires synergistic manipulation of multiple processing parameters. In addition to the substrate temperature and the rate of deposition, the angle of incidence of the deposition flux is known to affect structure evolution processes during film formation. However, its role in influencing the evolution of the intrinsic stress has not been determined. In this work, we investigate the effects of oblique-angle deposition on intrinsic stress evolution in polycrystalline gold and nickel films, using *in situ* stress measurements as well as *ex situ* surface and microstructure characterization. We find that as the angle between the surface normal and the direction of the deposition flux increases, the thickness at which film coalescence occurs increases and the post-coalescence stress shifts toward the tensile direction. We suggest that the first trend is associated with the effects of shadowing on the nucleation density in the pre-coalescence regime, and we attribute the second trend to an increase in surface roughness and shadowing effects associated with the dome-shaped surfaces of individual grains. According to this view, oblique-angle deposition on a mesoscopically rough surface eliminates or reduces condensation at and near grain boundaries, and therefore lowers the rate of adatom–grain boundary attachment, resulting in reduction in the compressive component of the intrinsic stress. The stress evolution map built from this work suggests routes to achieve specific stress levels in polycrystalline films.

Keywords: Intrinsic stress; Thin films; Shadowing; *In situ* measurement; Surface roughness

1. Introduction

The use of polycrystalline thin films is ubiquitous in emerging fields of science and technology, from microelectronics and flexible electronics to micro- and nano-manufacturing [1–5]. For many applications, control of the residual stress in the films remains a central but difficult task [6]. Residual stress includes extrinsic factors, such as thermal stress due to differential thermal expansion, and intrinsic factors, such as intrinsic stress caused by structure evolution during film deposition [7]. Intrinsic stresses can dominate in determining the residual stress in a film [1,7]. For films grown from the vapor phase, for example, intrinsic stresses of the order of 100 MPa or 1 GPa can be generated during the film formation processes [8,9]. Such high levels of stress can cause formation of voids and hillocks, cracking, buckling, delamination and other reliability problems [1,2,7]. The intrinsic stresses can also have profound effects on the properties and performance of films released from their substrates to perform mechanical functions as in nano- and micro-electromechanical devices such as nanobeam-based sensors and actuators [10,11].

The intrinsic stress in polycrystalline films is often correlated with surface and microstructure evolution processes during film formation [12–21]. Polycrystalline films form

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through nucleation of islands that grow to impinge and coalesce to form a continuous film. This is known as the Volmer–Weber mode of film formation [22]. Three types of intrinsic stress evolution behaviors have been identified during evaporative deposition of polycrystalline films [8,9,23]. In all three types of behaviors, a tensile stress develops during island coalescence due to an elastic deformation that occurs when islands meet and grain boundaries form through a "zipping" process [24,25]. After the film becomes continuous, the intrinsic stress continues to evolve in a tensile state under conditions of low atomic mobility (Type I behavior) or evolves toward or into a compressive state under conditions of high atomic mobility (Type II). Under conditions of intermediate atomic mobility, the evolution of post-coalescence stress involves a turnaround from a compressive state to a tensile state (intermediate behavior type) [23]. These stress evolution behaviors are controlled by a number of kinetic processes during film deposition, including adatom diffusion on the grain surface, adatom attachment to grain boundaries and grain growth during film thickening [16,23,26,27]. The homologous temperature T_h (defined as the substrate temperature divided by the melting temperature in K, i.e. $T_h = T/T_m$) and the rate of deposition are known to strongly influence these behaviors [8,9,16,23,28].

Precise control of the residual stress in thin films requires simultaneous manipulation of multiple processing parameters during deposition. In addition to the substrate temperature and deposition rate, the angle of the incident deposition flux is known to influence the surface morphology and grain structure of polycrystalline films through self-shadowing effects that lead to roughening of the film surface and vacancy formation in the bulk of the film [29–34]. However, the effects of the deposition angle on intrinsic stress evolution in thin films have not been investigated. As the first report of *in situ* stress measurements for oblique-angle deposition, we show here that the angle of the incident flux significantly affects the evolution of the intrinsic stress during growth of polycrystalline gold and nickel films. Specifically, a higher incidence angle leads to (i) an increase of the nominal thickness at which islands coalesce to form a continuous film (i.e. the coalescence thickness), and (ii) an overall less compressive or a more tensile post-coalescence stress in the film. We explain the first trend by self-shadowing in the pre-coalescence stage, and attribute the second trend to an increase in surface roughness and the shadowing effects associated with the dome-shaped surfaces of the grains in the post-coalescence stage. These findings lead to a stress evolution map in which the three types of stress evolution behaviors (i.e. Type I, intermediate type and Type II) are categorized as a function of the homologous temperature and the incidence angle.

2. Experimental methods

Gold and nickel films were deposited in an ultra-high-vacuum e-beam evaporation system, with a base pressure of 5.0×10^{-9} Torr. The deposition rate measured using a quartz crystal microbalance was adjusted to ensure that the real deposition flux (number of incident atoms per unit area of substrate surface per time) was the same for depositions carried out at different incident flux angles. All the films were deposited at a deposition rate normal to the plane of the substrate surface of 0.5 Å s^{-1}, as confirmed using atomic force microscopy (AFM) measurements. The surface and grain structures of the films were characterized using a Veeco Nanoscope IV AFM in tapping mode and a JEOL 2010 transmission electron microscope.

Intrinsic stress evolution during film growth was measured *in situ* using the cantilever deflection method. The details of this method can be found elsewhere [23,35,36]. To enable *in situ* measurements during oblique-angle deposition, we designed and fabricated a tiltable stress sensor platform, with which the inclination of the cantilever could be varied from 0° to 90°, as shown in the schematic illustration in Fig. 1a, where α refers to the angle of the incident flux relative to the direction normal to the substrate surface. A picture of our tiltable stress sensor setup is shown in Fig. 1b. It is important to note that the deflection of

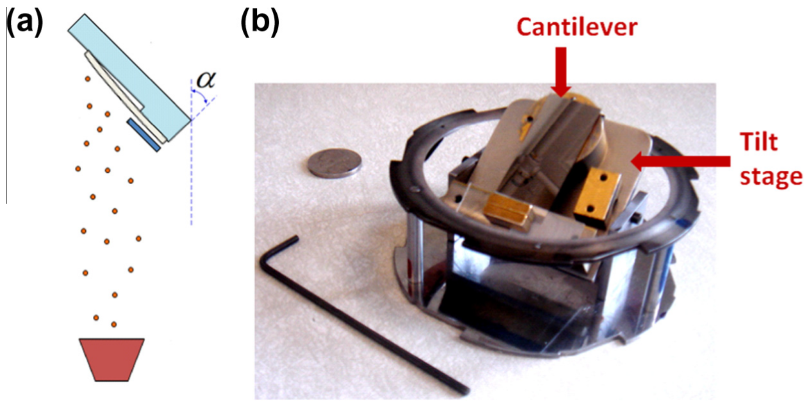

Fig. 1. (a) Illustration of the configuration used for *in situ* stress measurements during oblique angle-deposition and (b) a picture of the tiltable stress sensor.