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# Stress evolution during growth of 1-D island arrays: Kinetics and length scaling

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To explore the mechanisms controlling residual stress in thin films, we have measured the stress evolution during electrodeposition of Ni on lithographically patterned substrates with different pattern spacings and growth rates. Studying films with a controlled island geometry allows us to relate the stress (measured using wafer curvature) to the evolution of the morphology. We analyze the measurements with a model that focuses on the stress that develops where adjacent islands grow together to form new elements of grain boundary.


*Keywords:* Thin film; Residual stress; Kinetics; Electrodeposition

Residual stress in polycrystalline thin films is a persistent problem, since it can significantly reduce film performance or lead to failure [1]. A deeper understanding would enable it to be predicted and controlled.

Numerous studies have shown how the stress evolution depends on the material, processing conditions and evolving microstructure (many studies are reviewed in Refs. [2–4]). Films with low atomic mobility tend to grow in a state of tensile stress while films with higher mobility are compressive. Similarly, raising the growth temperature [5] or decreasing the growth rate [6] can change the stress from tensile to compressive. During growth, the stress goes through multiple states that correspond to the evolving microstructure, i.e. from isolated nuclei (low or compressive stress) through island coalescence (tensile stress) to a continuous film (steady-state stress that depends on growth rate) [7].

Various mechanisms have been proposed to explain different aspects of the stress evolution. Compressive stress in the nucleation stage has been attributed to lattice compression induced by the surface stress [8]. Hoffman [9] proposed that the tensile stress arises due to attractive forces between islands when the grain boundary forms, similar to the reason for crack closure [10]. The origin of the post-coalescence compressive stress is more controversial, with various groups attributing it to: adatoms on the surface [11], stress from the pre-coalescence stage [12], trapping of atoms between surface steps [13] and the insertion of atoms into the grain boundary during growth [14].

We have recently proposed a model [15] (described below) that focuses on the stress that develops at the point where layers in adjacent grains grow together to form new segments of grain boundary. Each layer's stress is predicted to depend on the rate at which the grain boundary height is changing when it forms. However, validation of this model is difficult because the shape of individual islands (and hence the rate of grain boundary formation) is not known during film growth. To overcome this problem, we have grown patterned arrays of islands in which the geometry during growth is known [16–18]. In the current work, we study stress evolution in a linear array in which the islands grow in the form of half-cylinders. Films with different pattern spacings ($L$) and growth rates ($R$) are measured to compare with the analytical model.

The samples consisted of Ni thin films electrodeposited on 100 μm thick Si substrates with 15 nm Ti and 150 nm Au layers deposited by electron beam evaporation on native oxide. After coating with photoresist, the substrate was lithographically patterned with linear trenches that exposed the Au underlayer for subsequent electrodeposition. Deposition was performed from a nickel sulfamate/

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boric acid bath; further details of the patterning and deposition process can be found in Ref. [17].

Substrates were prepared with trench arrays spaced 5.3, 10.6 or 26.5 µm apart, with corresponding trench widths of 2, 4 and 10 µm. Films were grown under potentiostatic control at multiple growth rates for each pattern spacing. Under these conditions, the growth rate normal to the surface is nearly constant so that each line grows into the form of a half-cylinder before it impinges on the one next to it. After the lines grow large enough to intersect (shown schematically in Fig. 1), they continue to grow at a constant radial growth rate. The inset shows a scanning electron microscopy (SEM) image of the morphology. The focused ion beam (FIB) cross-section shows that the shape of each island is semicircular and consists of multiple grains.

The evolution of the stress was measured using a real-time multibeam wafer curvature technique (MOS) [4] during deposition. To relate the measured curvature to the film stress, we must account for the fact that the patterned films are not spatially uniform or rotationally symmetric in the plane of the film. As shown in Figure 1, the film is assumed to consist of an array of islands that are semicircular in cross-section with radius $r$ spaced by $L$ in the $x$-direction. In the $y$-direction, the film is translationally symmetric. For $r > L/2$, the islands overlap so that a planar boundary forms between them, analogous to the grain boundary that forms in a polycrystalline film. We define the height of the boundary as $h_{gb}$. The curvature is measured along the $x$-direction ($\kappa_x$), i.e. normal to the interface that forms between adjacent lines.

We relate the measured curvature to the stress following the approach described in Freund and Suresh [19] for anisotropic films:

$$\kappa_x = \frac{6(f_x - \upsilon_s f_y)}{E_s h_s^2}, \tag{1}$$

where $E_s$, $\boldsymbol{v}_s$ and $h_s$ are the elastic modulus, Poisson's ratio and thickness of the substrate, respectively. $f_x$ and $f_y$ are due to the stress components in the film in the $x$- and $y$-direction, respectively. It is the deformation they induce plus compatibility that results in substrate curvature. For a uniform film, $f_x = f_y = \langle\boldsymbol{\sigma}\rangle h_f$, where $\langle\boldsymbol{\sigma}\rangle$ is the average equi-biaxial stress and $h_f$ is the film thickness. In this case, the typical Stoney's formula is recovered.

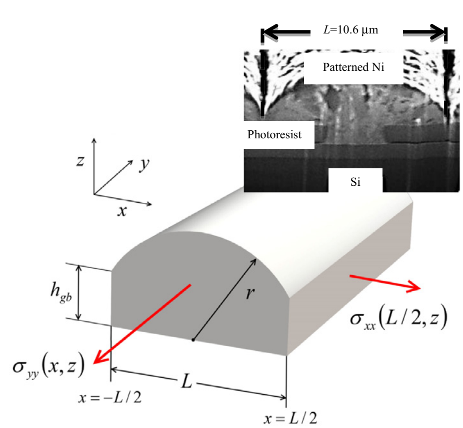


**Figure 1.** Schematic of island geometry used for analyzing stress evolution. (Inset) SEM image of Ni island array with front surface prepared by FIB cross-section.

The force in the $x$-direction ($f_x$) can be found by integrating the stress normal to the boundary between the lines ($\boldsymbol{\sigma}_{xx}$ at $x = L/2$) over the thickness of the film. This stress is independent of the $y$-position so that $f_x$ is given by:

$$f_x = \langle \sigma \rangle_{xx} h_f = \int_0^{h_{gb}} \sigma_{xx}(x = \frac{L}{2}, z) dz \tag{2}$$

Here we define $\langle\boldsymbol{\sigma}_{xx}\rangle$ as the average normal stress in the $x$-direction.

Similarly, we can compute $f_y$ due to stress in the $y$-direction along the length of the lines. Integrating over the cross-sectional area of the film gives:

$$f_y = \langle \sigma \rangle_{yy} h_f = \frac{1}{L} \int_0^{\infty} \int_{-L/2}^{L/2} \sigma_{yy}(x, z) dx dz, \tag{3}$$

where we define $\langle\boldsymbol{\sigma}_{yy}\rangle$ as the average normal stress in the $y$-direction.

Eq. (1) is valid when the thickness of the film is much less than the thickness of the substrate. When this is not the case, Freund et al. [20] have calculated a correction factor relating the measured curvature to the curvature expected from the Stoney equation. Although this formula was originally derived for a film that is spatially uniform and isotropic, we use it for the patterned films following Ref. [17]:

$$\kappa = \kappa_{ST} \left(1 + \frac{h_f}{h_s}\right) \left[1 + 4\frac{h_f}{h_s}\frac{\bar{E}_f}{\bar{E}_s} + 6\frac{h_f^2}{h_s^2}\frac{\bar{E}_f}{\bar{E}_s} + 4\frac{h_f^3}{h_s^3}\frac{\bar{E}_f}{\bar{E}_s} + \frac{h_f^4}{h_s^4}\frac{\bar{E}_f^2}{\bar{E}_s^2}\right]^{-1}. \tag{4}$$

$\bar{E}_f/\bar{E}_s$ is the ratio of the plane strain moduli for the film and substrate, which is taken here as 1.4 [17]. The measured curvature ($\kappa$) was divided by this correction factor to obtain the curvature that would be expected from a thin anisotropic film.

Measurements of the curvature vs. time are shown in Figure 2a–c for pattern spacings of 5.3, 10.6 and 26.5 µm, respectively. The growth rates for each measurement are indicated on the figure. The curvature has been multiplied by $E_s h_s^2/6$ so that it is equal to $(\langle\boldsymbol{\sigma}_{xx}\rangle - \nu_s \langle\boldsymbol{\sigma}_{yy}\rangle) h_f$, referred to as the stress-thickness.

The evolution of the stress-thickness has similar features for all of the measured conditions. When the radius is less than $L/2$, parallel islands do not make contact and the change in the stress-thickness is relatively small. After they start to impinge, the stress-thickness rises rapidly. Subsequently, the slope of the stress-thickness decreases until it reaches a steady state with a relatively constant slope. For the faster growth rates, the steady-state slope is positive (corresponding to tensile stress), while for slower growth rates the slope becomes less tensile or even compressive. This behavior is similar to that seen in polycrystalline unpatterned films, but the coalescence here occurs at much larger film thickness because the patterned island spacing is much larger than the typical grain size in unpatterned polycrystalline films.

In order to interpret these results, we use a model developed for stress in polycrystalline films [15]. It has been described previously so will only be discussed briefly here. The model is based on rate equations that describe the stress-generating processes that occur in each layer (indexed by the letter $i$) at the evolving grain boundary. We assume that the stress in individual layers in the film is independent of the stress in other layers (which Guduru refers to as a linear spring model [21]). The model contains two competing mechanisms of tensile and compressive stress generation.