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Multi-layer thin films/substrate system subjected to non-uniform misfit strains

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Abstract

Current methodologies used for the inference of thin film stress through curvature measurement are strictly restricted to stress and curvature states that are assumed to remain uniform over the entire film/substrate system. These methodologies have recently been extended to a single layer of thin film deposited on a substrate subjected to the non-uniform misfit strain in the thin film. Such methodologies are further extended to multi-layer thin films deposited on a substrate in the present study. Each thin film may have its own non-uniform misfit strain. We derive relations between the stresses in each thin film and the change of system curvatures due to the deposition of each thin film. The interface shear stresses between the adjacent films and between the thin film and the substrate are also obtained from the system curvatures. This provides the basis for the experimental determination of thin film stresses in multi-layer thin films on a substrate.

Keywords: Multi-layer thin films; Non-uniform misfit strain; Non-uniform wafer curvatures; Non-local stress-curvature relations; Interfacial shears

1. Introduction

Stoney (1909) studied a system composed of a thin film of thickness h_f, deposited on a relatively thick substrate, of thickness h_s, and derived a simple relation between the curvature, κ, of the system and the stress, $\sigma^{(f)}$, of the film as follows:

$$\sigma^{(f)} = \frac{E_s h_s^2 \kappa}{6h_f(1-\nu_s)}. \tag{1.1}$$

In the above the subscripts "f" and "s" denote the thin film and substrate, respectively, and E and ν are the Young's modulus and Poisson's ratio. Eq. (1.1) is called the Stoney formula, and it has been extensively used

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in the literature to infer film stress changes from experimental measurement of system curvature changes (Freund and Suresh, 2004).

Stoney's formula was based on the following assumptions, some of which have been relaxed.

(i) Both the film thickness h_f and the substrate thickness h_s are uniform and $h_f \ll h_s \ll R$, where R represents the characteristic length in the lateral direction (e.g., system radius R shown in Fig. 1). This assumption was recently relaxed for the thin film and substrate of different radii (Feng et al., 2006) and for arbitrarily non-uniform film thickness (Ngo et al., 2007). Their analytical results have been verified the X-ray microdiffraction experiments (Brown et al., 2007).

(ii) The strains and rotations of the plate system are infinitesimal. This assumption has been relaxed by various "large" deformation analyses (Masters and Salamon, 1993; Salamon and Masters, 1995; Finot et al., 1997; Freund, 2000), some of which have been validated by experiments (Lee et al., 2001; Park et al., 2003).

(iii) Both the film and substrate are homogeneous, isotropic, and linearly elastic. To our best knowledge this assumption has not been relaxed yet.

(iv) The film stress states are equi-biaxial (two equal stress components in any two, mutually orthogonal in-plane directions) while the out-of-plane direct stress and all shear stresses vanish. This assumption has been relaxed for non-equi-biaxial stress states (Shen et al., 1996; Wikstrom et al., 1999a; Park and Suresh, 2000; Freund and Suresh, 2004).

(v) The system's curvature components are equi-biaxial (two equal direct curvatures) while the twist curvature vanishes in all directions. This assumption has been relaxed for non-equi-biaxial curvature components and non-vanishing twist components (Shen et al., 1996; Wikstrom et al., 1999b; Park and Suresh, 2000; Freund and Suresh, 2004).

(vi) All surviving stress and curvature components are spatially constant over the plate system's surface, a situation that is often violated in practice. Recently, Huang et al. (2005) and Huang and Rosakis (2005) relaxed this assumption for the thin film/substrate system subjected to non-uniform, axisymmetric misfit strain (in thin film) and temperature change (in both thin film and substrate), respectively, while Ngo et al. (2006) and Huang and Rosakis (in press) studied the thin film/substrate system subject to arbitrarily non-uniform (e.g., non-axisymmetric) misfit strain and temperature. Their most important result is that *the film stresses depend non-locally on the system curvatures*, i.e., they depend on curvatures of the entire system.

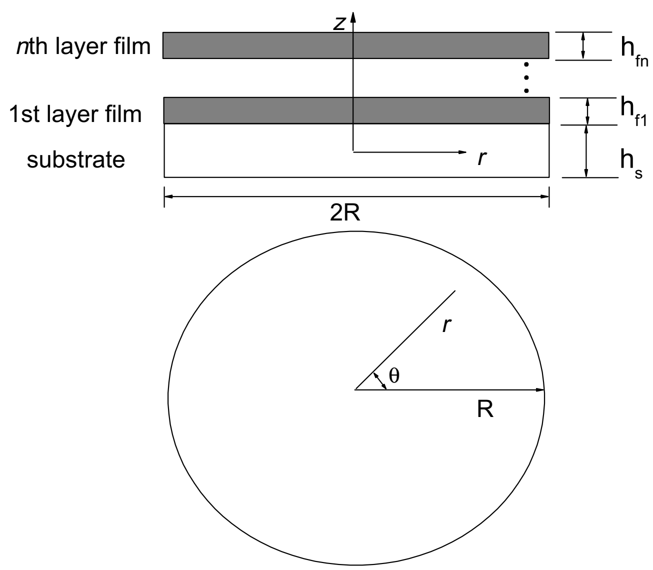

Fig. 1. A schematic diagram of multi-layer thin films deposited on a substrate, showing the cylindrical coordinates (r, θ, z).

Despite the explicitly stated assumptions of spatial stress and curvature uniformity, the Stoney formula is often, arbitrarily, applied to cases of practical interest where these assumptions are violated. This is typically done by applying Stoney's formula pointwise and thus extracting a local value of stress from a local measurement of the curvature of the system. This approach of inferring film stress clearly violates the uniformity assumptions of the analysis and, as such, its accuracy as an approximation is expected to deteriorate as the levels of curvature non-uniformity become more severe.

Many thin film/substrate systems involve multiple layers of thin films. The main purpose of this paper is to extend the above analyses by Huang, Rosakis and co-workers to a system composed of multi-layer thin films on a substrate subjected to non-uniform misfit strain distribution. We will relate stresses in each film and system curvatures to the misfit strain distribution, and ultimately derive a relation between the stresses in each film and system curvatures that would allow for the accurate experimental inference of film stresses from full-field and real-time curvature measurements.

2. Axisymmetric misfit strains

We first consider a system of multi-layer thin films deposited on a substrate subjected to axisymmetric misfit strain distribution $\varepsilon_m^{(i)}(r)$ in the ith layer ($i = 1, 2, \ldots, n$), where r is the radial coordinate, and n is the total number of layers of thin films (Fig. 1). The thin films and substrate are circular in the lateral direction and have a radius R. The deformation is axisymmetric and is therefore independent of the polar angle θ.

2.1. Governing equations

Let $h_{\mathrm{f}_i} (i = 1, 2, \ldots n)$ denote the thickness of the ith thin film (Fig. 1). The total thickness $h_\mathrm{f} = \sum_{i=1}^{n} h_{\mathrm{f}_i}$ of all n films is much less than the substrate thickness h_s, and both are much less than R, i.e. $h_\mathrm{f} \ll h_\mathrm{s} \ll R$. The Young's modulus and Poisson's ratio of the ith thin film and substrate are denoted by E_{f_i}, ν_{f_i}, E_s and ν_s, respectively.

The substrate is modeled as a plate since it can be subjected to bending and $h_\mathrm{s} \ll R$. The thin films are modeled as membranes that have no bending rigidities due to their small thickness $h_\mathrm{f} \ll h_\mathrm{s}$. Therefore they all have the same in-plane displacement $u_\mathrm{f}(r)$ in the radial (r) direction. The strains are $\varepsilon_{rr} = \frac{\mathrm{d}u_\mathrm{f}}{\mathrm{d}r}$ and $\varepsilon_{\theta\theta} = \frac{u_\mathrm{f}}{r}$. The stresses in the ith thin film can be obtained from the linear elastic constitutive model as

$$\begin{aligned} \sigma_{rr}^{(\mathrm{f}_i)} &= \frac{E_{\mathrm{f}_i}}{1-\nu_{\mathrm{f}_i}^2}\left[\frac{\mathrm{d}u_\mathrm{f}}{\mathrm{d}r} + \nu_{\mathrm{f}_i}\frac{u_\mathrm{f}}{r} - (1+\nu_{\mathrm{f}_i})\varepsilon_m^{(i)}\right], \\ \sigma_{\theta\theta}^{(\mathrm{f}_i)} &= \frac{E_{\mathrm{f}_i}}{1-\nu_{\mathrm{f}_i}^2}\left[\nu_{\mathrm{f}_i}\frac{\mathrm{d}u_\mathrm{f}}{\mathrm{d}r} + \frac{u_\mathrm{f}}{r} - (1+\nu_{\mathrm{f}_i})\varepsilon_m^{(i)}\right]. \end{aligned} \tag{2.1}$$

The membrane forces in the ith thin film are

$$N_r^{(\mathrm{f}_i)} = h_{\mathrm{f}_i}\sigma_{rr}^{(\mathrm{f}_i)}, \quad N_\theta^{(\mathrm{f}_i)} = h_{\mathrm{f}_i}\sigma_{\theta\theta}^{(\mathrm{f}_i)}. \tag{2.2}$$

For non-uniform misfit strain $\varepsilon_m^{(i)}(r)$, the shear stress tractions along the film/film and film/substrate interfaces do not vanish, and are denoted by $\tau^{(i)}(r)$ ($i = 1, 2, \ldots, n$) as shown in Fig. 2. The normal stress tractions still vanish because thin films have no bending rigidities. The equilibrium equations for thin films, accounting for the effect of interface shear stress tractions, become

$$\frac{\mathrm{d}N_r^{(\mathrm{f}_i)}}{\mathrm{d}r} + \frac{N_r^{(\mathrm{f}_i)} - N_\theta^{(\mathrm{f}_i)}}{r} - (\tau_i - \tau_{i+1}) = 0, \tag{2.3}$$

where $\tau_{n+1} = 0$ for the traction free surface. Substitution of Eqs. (2.1) and (2.2) into (2.3) and the summation of its left hand side yield

$$\left(\frac{\mathrm{d}^2u_\mathrm{f}}{\mathrm{d}r^2} + \frac{1}{r}\frac{\mathrm{d}u_\mathrm{f}}{\mathrm{d}r} - \frac{u_\mathrm{f}}{r^2}\right)\sum_{i=1}^{n}\frac{E_{\mathrm{f}_i}h_{\mathrm{f}_i}}{1-\nu_{\mathrm{f}_i}^2} = \tau^{(1)} + \sum_{i=1}^{n}\frac{E_{\mathrm{f}_i}h_{\mathrm{f}_i}}{1-\nu_{\mathrm{f}_i}}\frac{\mathrm{d}\varepsilon_m^{(i)}}{\mathrm{d}r}. \tag{2.4}$$

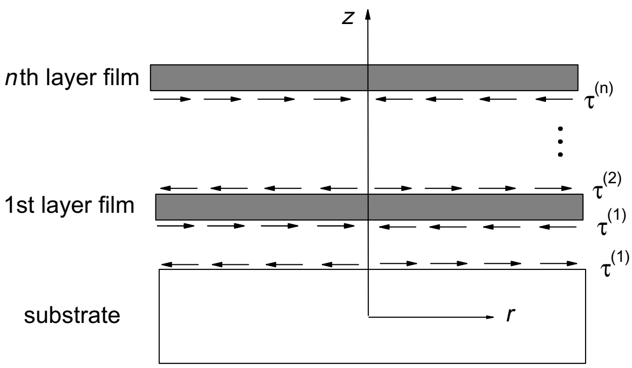

Fig. 2. A schematic diagram of the non-uniform shear traction distribution at the film/substrate and film/film interfaces.

Let u_s denote the displacement in the radial (r) direction at the neutral axis of the substrate, and w the displacement in the normal (z) direction. The forces and bending moments in the substrate are obtained from the linear thermo-elastic constitutive model as

$$N_r^{(s)} = \frac{E_s h_s}{1-\nu_s^2}\left(\frac{du_s}{dr} + \nu_s \frac{u_s}{r}\right),$$
$$N_\theta^{(s)} = \frac{E_s h_s}{1-\nu_s^2}\left(\nu_s\frac{du_s}{dr} + \frac{u_s}{r}\right), \tag{2.5}$$

$$M_r = \frac{E_s h_s^3}{12(1-\nu_s^2)}\left(\frac{d^2w}{dr^2} + \frac{\nu_s}{r}\frac{dw}{dr}\right),$$
$$M_\theta = \frac{E_s h_s^3}{12(1-\nu_s^2)}\left(\nu_s\frac{d^2w}{dr^2} + \frac{1}{r}\frac{dw}{dr}\right). \tag{2.6}$$

The shear stress $\tau^{(1)}$ at the thin film/substrate interface is equivalent to the distributed axial force $\tau^{(1)}$ and bending moment $\frac{h_s}{2}\tau^{(1)}$ applied at the neutral axis of the substrate. The in-plane force equilibrium equation of the substrate then becomes

$$\frac{dN_r^{(s)}}{dr} + \frac{N_r^{(s)} - N_\theta^{(s)}}{r} + \tau^{(1)} = 0. \tag{2.7}$$

The out-of-plane force and moment equilibrium equations are given by

$$\frac{dM_r}{dr} + \frac{M_r - M_\theta}{r} + Q - \frac{h_s}{2}\tau^{(1)} = 0, \tag{2.8}$$

$$\frac{dQ}{dr} + \frac{Q}{r} = 0, \tag{2.9}$$

where Q is the shear force normal to the neutral axis. Substitution of Eq. (2.5) into Eq. (2.7) yields

$$\frac{d^2u_s}{dr^2} + \frac{1}{r}\frac{du_s}{dr} - \frac{u_s}{r^2} = -\frac{1-\nu_s^2}{E_s h_s}\tau^{(1)}. \tag{2.10}$$

Elimination of Q from Eqs. (2.8) and (2.9), in conjunction with Eq. (2.6), gives

$$\frac{d^3w}{dr^3} + \frac{1}{r}\frac{d^2w}{dr^2} - \frac{1}{r^2}\frac{dw}{dr} = \frac{6(1-\nu_s^2)}{E_s h_s^2}\tau^{(1)}. \tag{2.11}$$

The continuity of displacement across the thin film/substrate interface requires

$$u_f = u_s - \frac{h_s}{2}\frac{dw}{dr}. \tag{2.12}$$

Eqs. (2.4) and (2.10), (2.11), (2.12) constitute four ordinary differential equations for u_f, u_s, w and $\tau^{(1)}$.

We can eliminate u_f, u_s and w from these four equations to obtain the shear stress $\tau^{(1)}$ at the thin film/substrate interface in terms of the misfit strains. For $h_f \ll h_s$, $\tau^{(1)}$ and the shear stresses $\tau^{(i)}$ $(i = 2, 3, \ldots, n)$ between thin films

$$\tau^{(i)} = -\sum_{j=i}^{n} \frac{E_{f_j} h_{f_j}}{1 - \nu_{f_j}} \frac{d\varepsilon_m^{(j)}}{dr}. \tag{2.13}$$

This is a remarkable result that holds regardless of boundary conditions at the edge $r = R$. Therefore, the interface shear stress is proportional to the gradient of misfit strains. For uniform misfit strains $\varepsilon_m^{(i)}(r) = \text{constant}$, the interface shear stress vanishes, i.e., $\tau^{(1)} = 0$.

Substitution of the above solution for shear stress $\tau^{(1)}$ into Eqs. (2.11) and (2.10) yields ordinary differential equations for displacements w and u_s in the substrate. Their solutions, at the limit of $h_f \ll h_s$, are

$$\frac{dw}{dr} = -6\frac{1-\nu_s^2}{E_s h_s^2}\frac{1}{r}\int_0^r \eta \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \varepsilon_m^{(i)}(\eta) d\eta + \frac{B_1}{2} r, \tag{2.14}$$

$$u_s = \frac{1-\nu_s^2}{E_s h_s}\frac{1}{r}\int_0^r \eta \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \varepsilon_m^{(i)}(\eta) d\eta + \frac{B_2}{2} r, \tag{2.15}$$

where B_1 and B_2 are to be determined. The displacement u_f in the film is obtained from the continuity condition (2.12) across the interface as

$$u_f = 4\frac{1-\nu_s^2}{E_s h_s}\frac{1}{r}\int_0^r \eta \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \varepsilon_m^{(i)}(\eta) d\eta + \left(\frac{B_2}{2} - \frac{h_s B_1}{4}\right) r. \tag{2.16}$$

The first boundary condition at the free edge $r = R$ requires that the net force vanish,

$$\sum_{i=1}^{n} N_r^{(f_i)} + N_r^{(s)} = 0 \text{ at } r = R, \tag{2.17}$$

which gives

$$B_2 = \frac{(1-\nu_s)^2}{E_s h_s} \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \overline{\varepsilon_m^{(i)}} \tag{2.18}$$

for $h_f \ll h_s$, where $\overline{\varepsilon_m^{(i)}} = \frac{2}{R^2}\int_0^R \eta \varepsilon_m^{(i)}(\eta) d\eta = \frac{\iint \varepsilon_m^{(i)} dA}{\pi R^2}$ is the average misfit strain in the ith thin film. The second boundary condition at the free edge $r = R$ is vanishing of net moment, i.e.,

$$M_r - \frac{h_s}{2}\sum_{i=1}^{n} N_r^{(f_i)} = 0 \text{ at } r = R, \tag{2.19}$$

which gives

$$B_1 = -6\frac{(1-\nu_s)^2}{E_s h_s^2} \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \overline{\varepsilon_m^{(i)}}. \tag{2.20}$$

2.2. Stresses in multi-layer thin films and system curvatures

The system curvatures are related to the out-of-plane displacement w by $\kappa_{rr} = \frac{d^2 w}{dr^2}$ and $\kappa_{\theta\theta} = \frac{1}{r}\frac{dw}{dr}$. The sum of these two curvatures is

$$\kappa_{rr} + \kappa_{\theta\theta} = -12\frac{1-\nu_s}{E_s h_s^2} \sum_{i=1}^{n} \frac{E_{f_i} h_{f_i}}{1-\nu_{f_i}} \left[\overline{\varepsilon_m^{(i)}} + \frac{1+\nu_s}{2}\left(\varepsilon_m^{(i)} - \overline{\varepsilon_m^{(i)}}\right)\right]. \tag{2.21}$$

The difference between two system curvatures is

$$\kappa_{rr} - \kappa_{\theta\theta} = -6\frac{1-\nu_s^2}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left[\varepsilon_m^{(i)} - \frac{2}{r^2}\int_0^r \eta\varepsilon_m^{(i)}(\eta)\mathrm{d}\eta\right]. \tag{2.22}$$

The sum and difference of stresses in each thin film are given by

$$\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)} = \frac{E_{f_i}}{1-\nu_{f_i}}\left(-2\varepsilon_m^{(i)}\right), \tag{2.23}$$

$$\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)} = 4\frac{E_{f_i}}{1+\nu_{f_i}}\frac{1-\nu_s^2}{E_s h_s}\sum_{j=1}^{n}\frac{E_{f_j}h_{f_j}}{1-\nu_{f_j}}\left[\varepsilon_m^{(j)} - \frac{2}{r^2}\int_0^r \eta\varepsilon_m^{(j)}(\eta)\mathrm{d}\eta\right]. \tag{2.24}$$

It is noted that $\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)}$ is in general expected to be smaller than $\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)}$ for $h_f/h_s \ll 1$.

2.3. Extension of Stoney formula for a multi-layer thin film/substrate system

We extend the Stoney formula for a multi-layer thin film/substrate system subjected to non-uniform misfits by establishing the direct relation between the stresses in each thin film and system curvatures. Both $\kappa_{rr} - \kappa_{\theta\theta}$ in Eq. (2.22) and $\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)}$ in Eq. (2.24) are proportional to $\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left[\varepsilon_m^{(i)} - \frac{2}{r^2}\int_0^r \eta\varepsilon_m^{(i)}(\eta)\mathrm{d}\eta\right]$. Elimination of misfit strains gives $\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)}$ in each film directly proportional to the difference $\kappa_{rr} - \kappa_{\theta\theta}$ in system curvatures,

$$\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)} = -\frac{2E_{f_i}h_s}{3(1+\nu_{f_i})}(\kappa_{rr} - \kappa_{\theta\theta}). \tag{2.25}$$

We now focus on the sum of thin-film stresses $\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)}$ and sum of system curvatures $\kappa_{rr} + \kappa_{\theta\theta}$. The average system curvature $\overline{\kappa_{rr} + \kappa_{\theta\theta}}$ is defined as

$$\overline{\kappa_{rr} + \kappa_{\theta\theta}} = \frac{1}{\pi R^2}\iint_A (\kappa_{rr} + \kappa_{\theta\theta})\eta\,\mathrm{d}\eta\,\mathrm{d}\theta = \frac{2}{R^2}\int_0^R \eta(\kappa_{rr} + \kappa_{\theta\theta})\mathrm{d}\eta. \tag{2.26}$$

It can be related to the average misfit strains by averaging both sides of Eq. (2.21), i.e.,

$$\overline{\kappa_{rr} + \kappa_{\theta\theta}} = -12\frac{1-\nu_s}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\overline{\varepsilon_m^{(i)}}. \tag{2.27}$$

The deviation from the average curvature, $\kappa_{rr} + \kappa_{\theta\theta} - \overline{\kappa_{rr} + \kappa_{\theta\theta}}$, can be related to the deviation from the average misfit strains as

$$\kappa_{rr} + \kappa_{\theta\theta} - \overline{\kappa_{rr} + \kappa_{\theta\theta}} = -6\frac{1-\nu_s^2}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left(\varepsilon_m^{(i)} - \overline{\varepsilon_m^{(i)}}\right). \tag{2.28}$$

Elimination of misfit strains $\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left(\varepsilon_m^{(i)} - \overline{\varepsilon_m^{(i)}}\right)$ and average misfit strains $\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\overline{\varepsilon_m^{(i)}}$ from Eqs. (2.27), (2.28) and (2.23) gives the sum of thin-film stresses in terms of system curvature as

$$\sum_{i=1}^{n}\frac{h_{f_i}}{h_f}\left(\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)}\right) = \frac{E_s h_s^2}{6(1-\nu_s)h_f}\left[\kappa_{rr} + \kappa_{\theta\theta} + \frac{1-\nu_s}{1+\nu_s}(\kappa_{rr} + \kappa_{\theta\theta} - \overline{\kappa_{rr} + \kappa_{\theta\theta}})\right], \tag{2.29}$$

where $h_f = \sum_{i=1}^{n}h_{f_i}$ is the total thickness of thin films. Eq. (2.29) only gives the weighted sum of stresses in all thin films, $\sum_{i=1}^{n}\frac{h_{f_i}}{h_f}\left(\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)}\right)$, in terms of the system curvatures, but not stresses in each thin film.

It is clear that the curvatures alone for a system with all n thin films are not sufficient to determine the stresses in all thin films. Additional parameters that can be measured in experiments are needed for the complete determination of all film stresses. One possibility is the system curvatures $\kappa_{rr}^{(i)}$ and $\kappa_{\theta\theta}^{(i)}$ after the first i thin films are deposited, and $\kappa_{rr}^{(n)} = \kappa_{rr}$ and $\kappa_{\theta\theta}^{(n)} = \kappa_{\theta\theta}$. The system curvatures $\kappa_{rr}^{(i)}$ and $\kappa_{\theta\theta}^{(i)}$ can be measured during the deposition process, or after the deposition process by etching the top $n-i$ thin films away. The stresses in the ith thin film are then given by

$$\sigma_{rr}^{(\mathrm{f}_i)} + \sigma_{\theta\theta}^{(\mathrm{f}_i)} = \frac{E_s h_s^2}{6(1-\nu_s)h_{\mathrm{f}_i}}\left[\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)} + \frac{1-\nu_s}{1+\nu_s}\left(\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)} - \overline{\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}}\right)\right], \tag{2.30}$$

where

$$\Delta\kappa_{rr}^{(i)} = \kappa_{rr}^{(i)} - \kappa_{rr}^{(i-1)} \text{ and } \Delta\kappa_{\theta\theta}^{(i)} = \kappa_{\theta\theta}^{(i)} - \kappa_{\theta\theta}^{(i-1)} \tag{2.31}$$

are the change of system curvatures due to the deposition of the ith thin film, and $\kappa_{rr}^{(0)} = \kappa_{\theta\theta}^{(0)} = 0$. The above equation is identical to its counterpart for a single layer of thin film (Ngo et al., 2006) except that its curvatures are replaced by the change of system curvatures $\Delta\kappa_{rr}^{(i)}$ and $\Delta\kappa_{\theta\theta}^{(i)}$ in Eq. (2.31). Eq. (2.31), together with Eq. (2.25), provides the direct relation between stresses in each thin film and system curvatures. The thin-film stresses at a point depend not only on the change of system curvatures $\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}$ at the same point (local dependence), but also on the average change of system curvatures $\overline{\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}}$ in the entire system (non-local dependence).

The interface stress $\tau^{(1)}$ between the substrate and the first thin film and $\tau^{(i)}$ between thin films in Eq. (2.13) can also be given by system curvatures

$$\tau^{(i)} = \frac{E_s h_s^2}{6(1-\nu_s^2)}\sum_{j=i}^{n}\frac{\mathrm{d}}{\mathrm{d}r}\left(\Delta\kappa_{rr}^{(j)} + \Delta\kappa_{\theta\theta}^{(j)}\right) = \frac{E_s h_s^2}{6(1-\nu_s^2)}\frac{\mathrm{d}}{\mathrm{d}r}\left(\kappa_{rr} + \kappa_{\theta\theta} - \kappa_{rr}^{(i-1)} - \kappa_{\theta\theta}^{(i-1)}\right), \tag{2.32}$$

where $\kappa_{rr}^{(0)} = \kappa_{\theta\theta}^{(0)} = 0$. The above equation provides a remarkably simple way to estimate the interface shear stresses from radial gradients of the two non-zero system curvatures. The shear stresses are responsible for promoting system failures through debonding of thin films.

3. Non-axisymmetric misfit strains

We extend the analysis in the previous section to a system of multi-layer thin films deposited on a substrate to arbitrary non-uniform misfit strains. The analysis is also an extension of Ngo et al. (2007) from a single thin film to multi-layer thin films. The non-uniform misfit strain in the ith thin film, $\varepsilon_m^{(i)}(r,\theta)$, can be expanded to the Fourier series

$$\varepsilon_m^{(i)}(r,\theta) = \sum_{k=0}^{\infty}\varepsilon_{kc}^{(i)}(r)\cos k\theta + \sum_{k=1}^{\infty}\varepsilon_{ks}^{(i)}(r)\sin k\theta, \tag{3.1}$$

where $\varepsilon_{0c}^{(i)}(r) = \frac{1}{2\pi}\int_0^{2\pi}\varepsilon_m^{(i)}(r,\theta)\mathrm{d}\theta$, $\varepsilon_{kc}^{(i)}(r) = \frac{1}{\pi}\int_0^{2\pi}\varepsilon_m^{(i)}(r,\theta)\cos k\theta\,\mathrm{d}\theta$ and $\varepsilon_{ks}^{(i)}(r) = \frac{1}{\pi}\int_0^{2\pi}\varepsilon_m^{(i)}(r,\theta)\sin k\theta\,\mathrm{d}\theta$ $(k \geqslant 1)$.

3.1. Stresses in multi-layer thin films and system curvatures

The system curvatures are

$$\kappa_{rr} = \frac{\partial^2 w}{\partial r^2}, \quad \kappa_{\theta\theta} = \frac{1}{r}\frac{\partial w}{\partial r} + \frac{1}{r^2}\frac{\partial^2 w}{\partial\theta^2}, \quad \kappa_{r\theta} = \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial w}{\partial\theta}\right). \tag{3.2}$$

The sum of system curvatures is related to the misfit strain by

$$\kappa_{rr} + \kappa_{\theta\theta} = -12\frac{1-\nu_s}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{\mathrm{f}_i}h_{\mathrm{f}_i}}{1-\nu_{\mathrm{f}_i}}\left\{\begin{array}{l}\overline{\varepsilon_m^{(i)}} + \frac{1+\nu_s}{2}\left(\varepsilon_m^{(i)} - \overline{\varepsilon_m^{(i)}}\right) \\ + \frac{1-\nu_s^2}{3+\nu_s}\sum\limits_{k=1}^{\infty}(k+1)\frac{r^k}{R^{2k+2}}\left[\begin{array}{l}\cos k\theta\int_0^R \eta^{k+1}\varepsilon_{kc}^{(i)}(\eta)\mathrm{d}\eta \\ +\sin k\theta\int_0^R \eta^{k+1}\varepsilon_{ks}^{(i)}(\eta)\mathrm{d}\eta\end{array}\right]\end{array}\right\}, \tag{3.3}$$

where $\overline{\varepsilon_m^{(i)}} = \frac{1}{\pi R^2}\iint_A \varepsilon_m^{(i)}(\eta,\varphi)\mathrm{d}A$ is the average misfit strain in the ith thin film, $\mathrm{d}A = \eta\,\mathrm{d}\eta\,\mathrm{d}\varphi$, and $\overline{\varepsilon_m^{(i)}}$ is also related to $\varepsilon_{0c}^{(i)}$ by $\overline{\varepsilon_m^{(i)}} = \frac{2}{R^2}\int_0^R \eta\varepsilon_{0c}^{(i)}(\eta)\mathrm{d}\eta$. The difference between two curvatures, $\kappa_{rr} - \kappa_{\theta\theta}$, and the twist $\kappa_{r\theta}$ are given by

$$\kappa_{rr} - \kappa_{\theta\theta} = -6\frac{1-\nu_s^2}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left\{\begin{array}{l} \varepsilon_m^{(i)} - \frac{2}{r^2}\int_0^r \eta\varepsilon_{0c}^{(i)}\mathrm{d}\eta \\ +\frac{1-\nu_s}{3+\nu_s}\sum_{k=1}^{\infty}\frac{k+1}{R^{k+2}}\left[k\frac{r^k}{R^k} - (k-1)\frac{r^{k-2}}{R^{k-2}}\right]\left(\begin{array}{l}\cos k\theta\int_0^R \eta^{k+1}\varepsilon_{kc}^{(i)}\mathrm{d}\eta \\ +\sin k\theta\int_0^R \eta^{k+1}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\end{array}\right) \\ -\sum_{k=1}^{\infty}\frac{k+1}{r^{k+2}}\left(\cos k\theta\int_0^r \eta^{k+1}\varepsilon_{kc}^{(i)}\mathrm{d}\eta + \sin k\theta\int_0^r \eta^{k+1}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\right) \\ -\sum_{k=1}^{\infty}(k-1)r^{k-2}\left(\cos k\theta\int_r^R \eta^{1-k}\varepsilon_{kc}^{(i)}\mathrm{d}\eta + \sin k\theta\int_r^R \eta^{1-k}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\right) \end{array}\right\}, \tag{3.4}$$

$$\kappa_{r\theta} = 3\frac{1-\nu_s^2}{E_s h_s^2}\sum_{i=1}^{n}\frac{E_{f_i}h_{f_i}}{1-\nu_{f_i}}\left\{\begin{array}{l} \frac{1-\nu_s}{3+\nu_s}\sum_{k=1}^{\infty}\frac{k+1}{R^{k+2}}\left[k\frac{r^k}{R^k} - (k-1)\frac{r^{k-2}}{R^{k-2}}\right]\left(\begin{array}{l}\sin k\theta\int_0^R \eta^{k+1}\varepsilon_{kc}^{(i)}\mathrm{d}\eta \\ -\cos k\theta\int_0^R \eta^{k+1}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\end{array}\right) \\ +\sum_{k=1}^{\infty}\frac{k+1}{r^{k+2}}\left(\sin k\theta\int_0^r \eta^{k+1}\varepsilon_{kc}^{(i)}\mathrm{d}\eta - \cos k\theta\int_0^r \eta^{k+1}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\right) \\ -\sum_{k=1}^{\infty}(k-1)r^{k-2}\left(\sin k\theta\int_r^R \eta^{1-k}\varepsilon_{kc}^{(i)}\mathrm{d}\eta - \cos k\theta\int_r^R \eta^{1-k}\varepsilon_{ks}^{(i)}\mathrm{d}\eta\right) \end{array}\right\}. \tag{3.5}$$

The sum $\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)}$ and differences $\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)}$ of stresses and the shear stress $\sigma_{r\theta}^{(f_i)}$ in the ith thin film are related to the misfit strains by

$$\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)} = \frac{E_{f_i}}{1-\nu_{f_i}}\left(-2\varepsilon_m^{(i)}\right), \tag{3.6}$$

$$\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)} = 4\frac{E_{f_i}}{1+\nu_{f_i}}\frac{1-\nu_s^2}{E_s h_s}\sum_{j=1}^{n}\frac{E_{f_j}h_{f_j}}{1-\nu_{f_j}}\left\{\begin{array}{l} \varepsilon_m^{(j)} - \frac{2}{r^2}\int_0^r \eta\varepsilon_{0c}^{(j)}\mathrm{d}\eta \\ -\sum_{k=1}^{\infty}\frac{k+1}{r^{k+2}}\left(\cos k\theta\int_0^r \eta^{k+1}\varepsilon_{kc}^{(j)}\mathrm{d}\eta + \sin k\theta\int_0^r \eta^{k+1}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\right) \\ -\sum_{k=1}^{\infty}(k-1)r^{k-2}\left(\cos k\theta\int_r^R \eta^{1-k}\varepsilon_{kc}^{(j)}\mathrm{d}\eta + \sin k\theta\int_r^R \eta^{1-k}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\right) \\ -\frac{\nu_s}{3+\nu_s}\sum_{k=1}^{\infty}\frac{k+1}{R^{k+2}}\left[k\frac{r^k}{R^k} - (k-1)\frac{r^{k-2}}{R^{k-2}}\right]\left(\begin{array}{l}\cos k\theta\int_0^R \eta^{k+1}\varepsilon_{kc}^{(j)}\mathrm{d}\eta \\ +\sin k\theta\int_0^R \eta^{k+1}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\end{array}\right) \end{array}\right\} \tag{3.7}$$

$$\sigma_{r\theta}^{(f_i)} = 2\frac{E_{f_i}}{1+\nu_{f_i}}\frac{1-\nu_s^2}{E_s h_s}\sum_{j=1}^{n}\frac{E_{f_j}h_{f_j}}{1-\nu_{f_j}}\left\{\begin{array}{l} -\sum_{k=1}^{\infty}\frac{k+1}{r^{k+2}}\left(\sin k\theta\int_0^r \eta^{k+1}\varepsilon_{kc}^{(j)}\mathrm{d}\eta - \cos k\theta\int_0^r \eta^{k+1}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\right) \\ +\sum_{k=1}^{\infty}(k-1)r^{k-2}\left(\sin k\theta\int_r^R \eta^{1-k}\varepsilon_{kc}^{(j)}\mathrm{d}\eta - \cos k\theta\int_r^R \eta^{1-k}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\right) \\ +\frac{\nu_s}{3+\nu_s}\sum_{k=1}^{\infty}\frac{k+1}{R^{k+2}}\left[k\frac{r^k}{R^k} - (k-1)\frac{r^{k-2}}{R^{k-2}}\right]\left(\begin{array}{l}\sin k\theta\int_0^R \eta^{k+1}\varepsilon_{kc}^{(j)}\mathrm{d}\eta \\ -\cos k\theta\int_0^R \eta^{k+1}\varepsilon_{ks}^{(j)}\mathrm{d}\eta\end{array}\right) \end{array}\right\}. \tag{3.8}$$

The shear stresses $\tau_r^{(1)}$ and $\tau_\theta^{(1)}$ between the first thin film and substrate and $\tau_r^{(i)}$ and $\tau_\theta^{(i)}$ $(i = 2, 3, \ldots, n)$ between thin films are related to the misfit strains by

$$\tau_r^{(i)} = -\sum_{j=i}^{n}\frac{E_{f_j}h_{f_j}}{1-\nu_{f_j}}\frac{\partial\varepsilon_m^{(j)}}{\partial r}, \quad \tau_\theta^{(i)} = -\sum_{j=i}^{n}\frac{E_{f_j}h_{f_j}}{1-\nu_{f_j}}\frac{1}{r}\frac{\partial\varepsilon_m^{(j)}}{\partial\theta}. \tag{3.9}$$

3.2. Extension of Stoney formula for a multi-layer thin film/substrate system

We extend the Stoney formula for a multi-layer thin film/substrate system by establishing the direct relation between the stresses in each thin film and system curvatures. We define the coefficients C_k and S_k in terms of the system curvatures $\kappa_{rr} + \kappa_{\theta\theta}$ by

$$\begin{aligned} C_k &= \frac{1}{\pi R^2}\iint_A (\kappa_{rr} + \kappa_{\theta\theta})\left(\frac{\eta}{R}\right)^k \cos k\varphi \, dA \\ S_k &= \frac{1}{\pi R^2}\iint_A (\kappa_{rr} + \kappa_{\theta\theta})\left(\frac{\eta}{R}\right)^k \sin k\varphi \, dA, \end{aligned} \tag{3.10}$$

where the integration is over the entire area A of the substrate, and $dA = \eta \, d\eta \, d\varphi$. The difference in stresses $\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)}$ and shear stress $\sigma_{r\theta}^{(f_i)}$ in the ith film are given in terms of system curvatures by

$$\sigma_{rr}^{(f_i)} - \sigma_{\theta\theta}^{(f_i)} = -\frac{E_{f_i} h_s}{6(1+\nu_{f_i})} \left\{ \begin{array}{l} 4(\kappa_{rr} - \kappa_{\theta\theta}) \\ -\sum_{k=1}^{\infty}(k+1)\left[k\left(\frac{r}{R}\right)^k - (k-1)\left(\frac{r}{R}\right)^{k-2}\right](C_k \cos k\theta + S_k \sin k\theta) \end{array} \right\}, \tag{3.11}$$

$$\sigma_{r\theta}^{(f_i)} = -\frac{E_{f_i} h_s}{6(1+\nu_{f_i})} \left\{ \begin{array}{l} 4\kappa_{r\theta} \\ +\frac{1}{2}\sum_{k=1}^{\infty}(k+1)\left[k\left(\frac{r}{R}\right)^k - (k-1)\left(\frac{r}{R}\right)^{k-2}\right](C_k \sin k\theta - S_k \cos k\theta) \end{array} \right\}, \tag{3.12}$$

Similar to Section 2.3, we define the system curvatures $\kappa_{rr}^{(i)}, \kappa_{\theta\theta}^{(i)}$ and $\kappa_{r\theta}^{(i)}$ after the first i thin films are deposited, which can be measured during the deposition process, or after the deposition process by etching the top $n-i$ thin films away. The changes of system curvatures due to the ith thin film are

$$\Delta\kappa_{rr}^{(i)} = \kappa_{rr}^{(i)} - \kappa_{rr}^{(i-1)}, \quad \Delta\kappa_{\theta\theta}^{(i)} = \kappa_{\theta\theta}^{(i)} - \kappa_{\theta\theta}^{(i-1)}, \quad \Delta\kappa_{r\theta}^{(i)} = \kappa_{r\theta}^{(i)} - \kappa_{r\theta}^{(i-1)}, \tag{3.13}$$

where $\kappa_{rr}^{(n)} = \kappa_{rr}$, $\kappa_{\theta\theta}^{(n)} = \kappa_{\theta\theta}$, $\kappa_{r\theta}^{(n)} = \kappa_{r\theta}$, and $\kappa_{rr}^{(0)} = \kappa_{\theta\theta}^{(0)} = \kappa_{r\theta}^{(0)} = 0$. We also define the coefficients $\Delta C_k^{(i)}$ and $\Delta S_k^{(i)}$ in terms of the changes of system curvatures $\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}$ by

$$\begin{aligned} \Delta C_k^{(i)} &= \frac{1}{\pi R^2}\iint_A \left(\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}\right)\left(\frac{\eta}{R}\right)^k \cos k\varphi \, dA \\ \Delta S_k^{(i)} &= \frac{1}{\pi R^2}\iint_A \left(\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}\right)\left(\frac{\eta}{R}\right)^k \sin k\varphi \, dA. \end{aligned} \tag{3.14}$$

The sum of stresses in the ith film is given in terms of the changes of system curvatures by

$$\sigma_{rr}^{(f_i)} + \sigma_{\theta\theta}^{(f_i)} = \frac{E_s h_s^2}{6h_{f_i}(1-\nu_s)} \left[\begin{array}{l} \Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)} + \frac{1-\nu_s}{1+\nu_s}\left(\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)} - \overline{\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}}\right) \\ -\frac{1-\nu_s}{1+\nu_s}\sum_{k=1}^{\infty}(k+1)\left(\frac{r}{R}\right)^k\left(\Delta C_k^{(i)} \cos k\theta + \Delta S_k^{(i)} \sin k\theta\right) \end{array} \right], \tag{3.15}$$

where $\overline{\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}} = \Delta C_0^{(i)} = \frac{1}{\pi R^2}\iint_A \left(\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}\right) dA$ is the average over entire area A of the substrate. Eqs. (3.11), (3.12) and (3.15) provide direct relations between stresses in thin films and system curvatures. It is important to note that stresses at a point in the thin film depend not only on curvatures at the same point (local dependence), but also on the curvatures in the entire substrate (non-local dependence).

The interface shear stresses $\tau_r^{(i)}$ and $\tau_\theta^{(i)}$ can also be directly related to substrate curvatures via

$$\tau_r^{(i)} = \frac{E_s h_s^2}{6\left(1-\nu_s^2\right)} \sum_{j=i}^{n} \left[\begin{array}{l} \frac{\partial}{\partial r}\left(\Delta\kappa_{rr}^{(j)} + \Delta\kappa_{\theta\theta}^{(j)}\right) \\ -\frac{1-\nu_s}{2R} \sum_{k=1}^{\infty} k(k+1)\left(\Delta C_k^{(j)} \cos k\theta + \Delta S_k^{(j)} \sin k\theta\right)\left(\frac{r}{R}\right)^{k-1} \end{array} \right]$$
$$= \frac{E_s h_s^2}{6(1-\nu_s^2)} \left\{ \begin{array}{l} \frac{\partial}{\partial r}\left(\kappa_{rr} + \kappa_{\theta\theta} - \kappa_{rr}^{(i-1)} - \kappa_{\theta\theta}^{(i-1)}\right) \\ -\frac{1-\nu_s}{2R} \sum_{k=1}^{\infty} k(k+1)\left[\left(\sum_{j=i}^{n} \Delta C_k^{(j)}\right) \cos k\theta + \left(\sum_{j=i}^{n} \Delta S_k^{(j)}\right) \sin k\theta\right]\left(\frac{r}{R}\right)^{k-1} \end{array} \right\}, \tag{3.16}$$

$$\tau_\theta^{(i)} = \frac{E_s h_s^2}{6(1-\nu_s^2)} \sum_{j=i}^{n} \left[\begin{array}{l} \frac{1}{r}\frac{\partial}{\partial \theta}\left(\Delta\kappa_{rr}^{(j)} + \kappa_{\theta\theta}^{(j)}\right) \\ +\frac{1-\nu_s}{2R} \sum_{k=1}^{\infty} k(k+1)\left(\Delta C_k^{(j)} \sin k\theta - \Delta S_k^{(j)} \cos k\theta\right)\left(\frac{r}{R}\right)^{k-1} \end{array} \right]$$
$$= \frac{E_s h_s^2}{6(1-\nu_s^2)} \left\{ \begin{array}{l} \frac{1}{r}\frac{\partial}{\partial \theta}\left(\kappa_{rr} + \kappa_{\theta\theta} - \kappa_{rr}^{(i-1)} - \kappa_{\theta\theta}^{(i-1)}\right) \\ +\frac{1-\nu_s}{2R} \sum_{k=1}^{\infty} k(k+1)\left[\left(\sum_{j=i}^{n} \Delta C_k^{(j)}\right) \sin k\theta - \left(\sum_{j=i}^{n} \Delta S_k^{(j)}\right) \cos k\theta\right]\left(\frac{r}{R}\right)^{k-1} \end{array} \right\}. \tag{3.17}$$

These provide a way to estimate the interface shear stresses from the gradients of system curvatures. They also display a non-local dependence.

4. Concluding remarks and discussion

The Stoney formula is extended in the present analysis for multi-layer thin films deposited on a substrate subjected to non-uniform misfit strains. For multi-layer thin films ($i = 1, 2, \ldots, n$) on a substrate, the total system curvature $\kappa_{rr} + \kappa_{\theta\theta}$ only gives the average stresses in all thin films, not stresses in each thin film. In the present study the stresses in the ith thin film are obtained in terms of the change of system curvatures $\Delta\kappa_{rr}^{(i)} + \Delta\kappa_{\theta\theta}^{(i)}$ due to the deposition of the ith thin film. The interface shear stresses between adjacent thin films and between the thin film and substrate are also obtained from the curvatures. This provides the basis for experimental determination of the stresses in each thin film and interface shear stresses.

Similar to a single layer of thin film on a substrate, the stresses in multi-layer thin films are related to the system curvatures, and such dependence is non-local since the stresses at a point on the film depend on both the local value of the system curvatures (at the same point) and on the value of curvatures of all other points on the plate system (non-local dependence).

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