Notes: ATG instability in thin film assemblies

0.1 A linear stability analysis assuming volume diffusion

Let us examine the stability of a single misfitting plate in the absence of any applied stresses. Let the initially flat interface $y_I = \pm\frac{h}{2}$ of the plate be perturbed sinusiodally (and symmetrically) as follows:

$$y_I(x) = \pm\left[\frac{h}{2} + \delta\cos(kx)\right], \tag{1}$$

where δ is the perturbation amplitude, k is the perturbation wave number, related to the perturbation wavelength λ by $k = 2\pi/\lambda$, and h is the thickness of the film (See the schematic 1). Assuming bulk diffusion to be the dominant diffusion mechanism, we would like to identify the wavenumbers for which the perturbation grows, and the growth rate for these wavenumbers as well. We closely follow Sridhar et al [1] in the following analysis; the present analysis is identical to theirs save the assumed diffusion mechanism: while Sridhar et al assume the perturbations to grow via interfacial diffusion, we assume that they grow via volume diffusion.
The evolution of the interface will be determined by the gradient in the diffusion potential (per atom) Φ. This diffusion potential at the interface depends on the local curvature of the interface and the local elastic stress and strain fields [2]:

$$\Phi = V_a\left(-\chi\gamma + [W]_p^m - \mathbf{T}\cdot[\nabla\mathbf{u}\cdot\mathbf{n}]_p^m\right), \tag{2}$$

where V_a is the atomic volume, χ is twice the mean curvature (or, in this case the local curvature of the interface itself) with respect to the p phase, $[q]_p^m$ is the jump in the quantity q across the interface , i.e., $(q)^m - (q)^p$, $\mathbf{T}$ is the traction at the interface, and, W is the strain energy density. In terms of stresses and strains (assuming the stress-free matrix to be the reference state), $[W]_p^m$ and $\mathbf{T}\cdot[\nabla\mathbf{u}\cdot\mathbf{n}]_p^m$ are written as follows:

$$[W]_p^m = \frac{1}{2}\left[\sigma^m\varepsilon^m - \sigma^p(\varepsilon^p - \varepsilon^T)\right]; \tag{3}$$

$$\mathbf{T}\cdot[\nabla\mathbf{u}\cdot\mathbf{n}]_p^m = \sigma^m[\varepsilon^p - \varepsilon^m]. \tag{4}$$

Thus, given an interface profile, we need to evaluate the local curvature and local elastic fields to study its morphological evolution. Given an interface profile $y(x)$, the local curvature χ is given by (See, for example [3]),

$$\chi = \frac{y''}{[1+(y')^2]^{\frac{3}{2}}}, \tag{5}$$

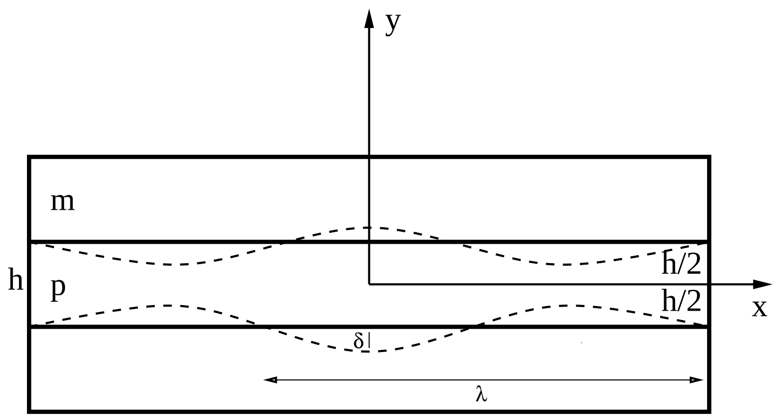

Figure 1: Schematic of the perturbation of the thin film assembly

where the $'$ denotes the differentiation of y with respect to x. Thus, for the assumed sinusoidal interface profile, to first order in δk, the curvature is given by

$$\chi \simeq -\delta k^2 \cos(kx). \tag{6}$$

On the other hand, in general, given an interface profile, it is not possible to obtain the analytical expressions for the elastic fields. However, since we are performing only the linear stability analysis, it is possible evaluate the displacement fields (and hence the stresses and strains) to first order in δk as shown below.
To evaluate the elastic fields for the given interface profile, we make the following assumptions:

1. Both the misfitting plate and the matrix are isotropic,
2. $\delta k \ll 1$ (so that the corrections to the elastic fields that arise due to the shape perturbation are kept only to first order in δk),
3. Poisson's ratio for both the phases are the same, and in particular is equal to $\frac{1}{3}$, and
4. The elastic problem is a plane strain problem.

The equations of equilibrium for the displacements $\mathbf{u}$ (in both the matrix and precipitate phases, for isotropic elasticity) are [4]

$$2(1-\nu)\nabla\nabla\cdot\mathbf{u} - (1-2\nu)\nabla\times\nabla\times\mathbf{u} = 0, \tag{7}$$

where ν is the Poisson's ratio.
Let the displacement solution $\mathbf{u}$ to the equation of mechanical equilibrium (7) be split as $\mathbf{u}^0 + \delta k \mathbf{u}^1$, where $\mathbf{u}^0$ is the solution for a plate with flat interfaces and $\mathbf{u}^1$ is the correction due to the shape perturbation. We assume the boundary between the plate and the matrix to be coherent, and hence, the displacement $\mathbf{u}^0$ and the traction $\mathbf{T}^0$ fields have to satisfy the following continuity conditions (See equation 6.3 on p. 39 of [5]):

$$[\mathbf{u}^0] = [\mathbf{T}^0] = 0 \;\; on \;\; y = \pm\frac{h}{2} \tag{8}$$

The displacement fields $\mathbf{u}^0$ that satisfy the partial differential equations (7) and the boundary conditions (8) are as follows:

$$\mathbf{u}^0 = \hat{\mathbf{y}}\frac{(1+\nu)\varepsilon^T}{(1-\nu)}y \ \ (within\ the\ plate); \tag{9}$$

$$\mathbf{u}^0 = \hat{\mathbf{y}}\frac{(1+\nu)\varepsilon^T}{(1-\nu)}\left(\frac{h}{2}\right) \ \ (within\ the\ matrix); \tag{10}$$

where, $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the unit normal vectors, ε^T is the dilatational eigenstrain.
The perturbation fields $\mathbf{u}^1$ satisfying the equations (7) should also satisfy the following boundary conditions:

1. By symmetry, in the plate (at $y = 0$), the y displacement component (u_y^1) and the shear stress component (σ_{xy}^1) should vanish, i.e.,

$$u_y^1 = \sigma_{xy}^1 = 0 \ \text{ at } \ y = 0; \tag{11}$$

2. Both components of the perturbation fields should vanish far away from the interface, well into the matrix phase, i.e.,

$$\mathbf{u}^1 \to 0 \ \ as \ \ y \to \infty. \tag{12}$$

The displacement field solutions to the equations (7) satisfying the above two conditions are the following:

$$\begin{aligned} \mathbf{u}^1 = & \\ & \hat{\mathbf{x}}[P_1 c(Y) + P_2 Y s(Y)]\sin(X) \\ & +\hat{\mathbf{y}}\{\ [(3-4\nu)P_2 - P_1]s(Y) - P_2 Y c(Y)\ \}\cos(X) \\ & (within\ the\ plate); \end{aligned} \tag{13}$$

and,

$$\begin{aligned} \mathbf{u}^1 = & \\ & \hat{\mathbf{x}}[M_1 - M_2 Y]\exp(-Y)\sin(X) \\ & +\hat{\mathbf{y}}[(M_1 - (3-4\nu)M_2) - M_2 Y]\exp(-Y)\cos(X) \\ & (within\ the\ matrix); \end{aligned} \tag{14}$$

where, $X = kx$, $Y = ky$, $c = \cosh$, and $s = \sinh$. The constants P_1, P_2, M_1, and M_2 are evaluated as follows:

- Apply the traction and displacement continuity conditions across the perturbed interface, i.e.,

$$[\mathbf{u}] = [\mathbf{T}] = 0 \ \ on \ \ y = \pm\left[\frac{h}{2} + \delta\cos(kx)\right]. \tag{15}$$

- Expand the above equations to first order in δk and match the coefficients of the first-order term in δk.

We note that for calculating the traction, we need to evaluate the unit normal vector to the interface. For the given interface expression, viz., $y = \pm\left[\frac{h}{2} + \delta\cos(kx)\right]$, the components of the unit normal vector (evaluated to first order in δk) are $n_x \simeq \delta k \sin(kx)$, and $n_y \simeq 1$ [6].
The procedure outlined above results in the following system of equations:

$$\begin{pmatrix} -c(\omega) & -\omega s(\omega) & (c(\omega)-s(\omega)) & (s(\omega)-c(\omega))\omega \\ s(\omega) & \omega c(\omega)-\frac{5}{3}s(\omega) & (c(\omega)-s(\omega)) & \frac{1}{3}(s(\omega)-c(\omega))(5+3\omega) \\ c(\omega) & \frac{1}{3}(3\omega s(\omega)-4c(\omega)) & \frac{(1-\aleph)}{(1+\aleph)}(s(\omega)-c(\omega)) & \frac{1}{3}\frac{(1-\aleph)}{(1+\aleph)}(c(\omega)-s(\omega))(4+3\omega) \\ s(\omega) & \frac{1}{3}(3\omega c(\omega)-s(\omega)) & \frac{(1-\aleph)}{(1+\aleph)}(c(\omega)-s(\omega)) & \frac{1}{3}\frac{(1-\aleph)}{(1+\aleph)}(s(\omega)-c(\omega))(1+3\omega) \end{pmatrix} \begin{pmatrix} P_1 \\ P_2 \\ P_3 \\ P_4 \end{pmatrix} = \begin{pmatrix} 0 \\ \frac{2\varepsilon^T}{k} \\ 0 \\ \frac{2\varepsilon^T}{k} \end{pmatrix},$$

where, $\aleph = \frac{(E_p - E_m)}{(E_p + E_m)}$, $\omega = \frac{kh}{2}$, and E_p, and E_m are the Young's moduli of the precipitate and matrix phases, respectively. By solving the above system of equations for the constsnts, we can obtain the correction to the displcement fields. From the displaecment fields, we can obtain the other elastic fileds, and hence $[W]_p^m$, and $\mathbf{T} \cdot [\nabla \mathbf{u} \cdot \mathbf{n}]_p^m$. The algebra involved in obtaining the last two terms in the diffusion potential expression is thus very cumbersome, and we have found MAPLE$^{\text{TM}}$ [7] to be extremely helpful in carrying out the same.

Having obtained the diffusion potential Φ, we assume that the atomic flux J is related to the diffusion potential linearly through the mobility M:

$$J = -M \nabla \Phi. \tag{16}$$

Since we are assuming the mass trnsport to occur via volume diffusion, the normal velocity v_i of the interface will be given by the following expression (See equation 11 of [8], for example):

$$v_i = M V_a \nabla \Phi. \tag{17}$$

However, the normal velocity of the interface is related to the interface profile through (See equation 6 of [1])

$$v_i \simeq \frac{\partial \delta}{\partial t} \cos(kx). \tag{18}$$

Equating the right-hand sides of the above two expressions for the interface velocity, and solving the resultant equation, we obtain

$$\delta(\tau) = \delta(0) \exp(\phi \tau), \tag{19}$$

where, $\tau = \frac{t V_a^2 M \gamma}{h^3}$ the dimensionless time and $\phi = \frac{G h^3}{M \gamma V_a^2}$ is the parameter that indicates the rate of growth (with G being the unnormalized value).

The growth rate ϕ can be written as follows:

$$\phi = \phi_c + \phi_e = -(kh)^3 + \phi_e, \tag{20}$$

where, ϕ_c and ϕ_e correspond to the curvature and elastic contributions to ϕ. As noted earlier, the calculation of ϕ_e, though cumbersome, can be obtained using MAPLE$^{\text{TM}}$ (see Section 0.2 for the details), and turns out to be

$$\phi_e = (kh)^2 \theta f(\alpha, \omega, s(\omega), c(\omega)), \tag{21}$$

where,

$$\theta = E_p h {\varepsilon^T}^2 / \gamma \tag{22}$$

with γ being the interfacial energy, and

$$f = \frac{A}{B} + \frac{18\exp(-\omega)(1-\alpha)\alpha c(\omega\omega)}{7s(\omega)c(\omega)\alpha^2 + 8c(\omega)s(\omega) + 4\alpha^2 + 3\alpha 2\omega - 8c(\omega)^2\alpha^2 + 8cosh(\omega)^2 + 12\omega\alpha - 4} - \frac{1.5c(\omega)^2\alpha(\alpha+4)}{7s(\omega)c(\omega)\alpha^2 + 8c(\omega)s(\omega) + 4\alpha^2 + 3\alpha 2\omega - 8c(\omega)^2\alpha^2 + 8cosh(\omega)^2 + 12\omega\alpha - 4} - \frac{3(12\omega s(\omega)\alpha + 3\omega s(\omega)\alpha^2 - 4c(\omega) - 6c(\omega)\alpha^2 - 4s(\omega) + 4s(\omega)\alpha^2 - 10c(\omega)\alpha)c(\omega)}{7s(\omega)c(\omega)\alpha^2 + 8c(\omega)s(\omega) + 4\alpha^2 + 3\alpha 2\omega - 8c(\omega)^2\alpha^2 + 8cosh(\omega)^2 + 12\omega\alpha - 4} + \frac{9c(\omega)\alpha(\alpha+4)\omega s(\omega)}{7s(\omega)c(\omega)\alpha^2 + 8c(\omega)s(\omega) + 4\alpha^2 + 3\alpha 2\omega - 8c(\omega)^2\alpha^2 + 8cosh(\omega)^2 + 12\omega\alpha - 4} + \frac{24\exp(-\omega)c(\omega)(1-\alpha)\alpha}{7s(\omega)c(\omega)\alpha^2 + 8c(\omega)s(\omega) + 4\alpha^2 + 3\alpha 2\omega - 8c(\omega)^2\alpha^2 + 8cosh(\omega)^2 + 12\omega\alpha - 4}$$

with,

$$A = 6\exp(-\omega)(1-\alpha)c(\omega)(-2c(\omega) - 2s(\omega) - 3c(\omega)\alpha + 2s(\omega)\alpha + 3\omega s(\omega)\alpha - 3\omega c(\omega)\alpha)$$

and,

$$B = 15c(\omega)^2 s(\omega)^2 - 4s(\omega)\alpha^2 - 3\omega s(\omega)\alpha^2 - 12\omega s(\omega)\alpha + 4s(\omega) + 11c(\omega)\alpha^2 - 15c(\omega)^3\alpha^2 + 4c(\omega) + 3c\omega\alpha^2\omega + 12\omega c(\omega)\alpha$$

Thus, while growth via surface diffusion gives rise to a curvature contribution of $(kh)^4$ and elastic contribution of $(kh)^3$, growth via volume diffusion results in a curvature contribution of $(kh)^3$ and elastic contribution of $(kh)^2$ to ϕ. These results are consistent with the fact that in the absence of elastic stress effects, the curvature contribution is $(kh)^4$ and $(kh)^3$ for surface and volume diffusion mechanisms respectively [9].
Similiar analysis can also be performed for an asymmetrically perturbed interface: i.e., $y_I = \pm\left(\frac{h}{2}\right) + \delta\cos(kx)$.

0.2 MAPLE$^{\text{TM}}$ *commands*

The following MAPLE$^{\text{TM}}$ commands are used to obtain the expression for the growth rate. A note on the symbols: $E[A] = E_p$; $E[B] = E_m$; and, $\eta = \epsilon^T$.

```
u[A] := delta*k*(P[1]*cosh(k*y) + P[2]*k*y*sinh(k*y))*sin(k*x);
v[A] := (1+nu)*y*eta/(1-nu)+delta*k*(((3-4*nu)*P[2]-P[1])*sinh(k*y) -
P[2]*k*y*cosh(k*y))*cos(k*x);
epsilon[1] := diff(u[A],x);
epsilon[2] := diff(v[A],y);
epsilon[3] := 0.5*(diff(u[A],y) + diff(v[A],x));
```

```
sigma[1] := E[A]/((1+nu)*(1-2*nu))*((1-nu)*(epsilon[1]-eta*(1+nu)) +
nu*(epsilon[2]-eta*(1+nu)));
sigma[2] := E[A]/((1+nu)*(1-2*nu))*((1-nu)*(epsilon[2]-eta*(1+nu)) +
nu*(epsilon[1]-eta*(1+nu)));
sigma[3] := E[A]/(1+nu) *epsilon[3];
w[1] := simplify((sigma[1]*(epsilon[1]-eta) + sigma[2]*(epsilon[2]-eta)
+ 2*sigma[3]*epsilon[3])/2);
u[B] := delta*k*(M[1]-M[2]*k*y)*exp(-k*y)*sin(k*x);
v[B] := h*(1+nu)*eta/(2*(1-nu)) + delta*k*((M[1]
-(3-4*nu)*M[2])-M[2]*k*y)*exp(-k*y)*cos(k*x);
epsilon[4] := diff(u[B],x);
epsilon[5] := diff(v[B],y);
epsilon[6] := 0.5*(diff(u[B],y) + diff(v[B],x));
sigma[4] := E[B]/((1+nu)*(1-2*nu))*((1-nu)*(epsilon[4]) +
nu*(epsilon[5]));
sigma[5] := E[B]/((1+nu)*(1-2*nu))*((1-nu)*(epsilon[5]) +
nu*(epsilon[4]));
sigma[6] := E[B]/(1+nu) *epsilon[6];
w[2] := simplify((sigma[4]*epsilon[4] + sigma[5]*epsilon[5] +
2*sigma[6]*epsilon[6])/2);
w[3] := simplify(sigma[4]*(epsilon[4]-epsilon[1]) +
sigma[5]*(epsilon[5]-epsilon[2]) + sigma[6]*(epsilon[6]-epsilon[3]));
w[4] := simplify(-Einterfacial*delta*k*k*cos(k*x) + w[1] - w[2] + w[3]);
w[5] := diff(w[4],x);
w[6] := diff(w[5],x);
with(linalg):
U := matrix(4,4,[-cosh(omega), -omega*sinh(omega),
(cosh(omega)-sinh(omega)),(sinh(omega)-cosh(omega))*omega,sinh(omega),
omega*cosh(omega)-5*sinh(omega)/3,(cosh(omega)-sinh(omega)),
(sinh(omega)-cosh(omega))*(5+3*omega)/3,cosh(omega),
(3*omega*sinh(omega)-4*cosh(omega))/3,
(1-alpha)*(sinh(omega)-cosh(omega))/(1+alpha),
(1-alpha)*(cosh(omega)-sinh(omega))*(4+3*omega)/(3*(1+alpha)),
sinh(omega),(3*omega*cosh(omega)-sinh(omega))/3,
(1-alpha)*(cosh(omega)-sinh(omega))/(1+alpha),
(1-alpha)*(sinh(omega)-cosh(omega))*(1+3*omega)/(3*(1+alpha))]);
V := matrix(4,1,[0,2.*eta/k,0,2*eta/k]);
Uinv := inverse(U);
PM := evalm(Uinv &* V);
P[1] := simplify(PM[1,1]);
P[2] := simplify(PM[2,1]);
M[1] := simplify(PM[3,1]);
M[2] := simplify(PM[4,1]);
w[7] := taylor(w[6],delta=0,2);
w[8] := subs(nu=1./3.,y=omega/k,E[B] = E[A]*(1-alpha)/(1+alpha),w[7]);
```

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