Asaro-Tiller-Grinfeld Instability

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1. Introduction

Asaro-Tiller-Grinfeld (ATG) Instability refers to the instability at the interface of solids or a solid and its melt/vapour. When the interface is experiencing some non-hydrostatic stress, the system may tend to undergo some morphological change that would relieve the elastic energy, thereby reducing the energy of the system. This instability was first predicted by Asaro and Tiller and then later independently rediscovered by Grinfeld and hence the name.

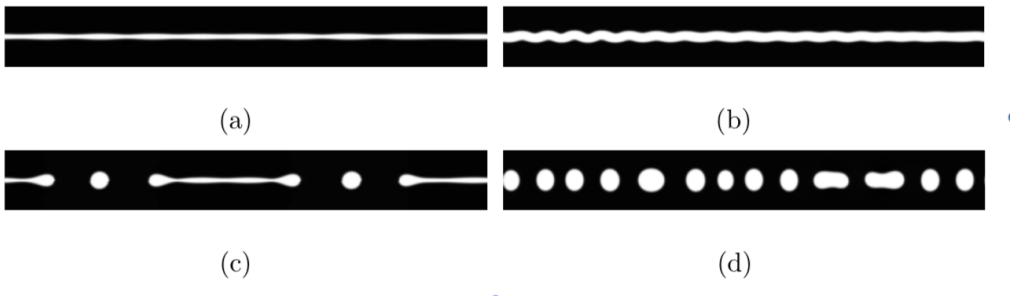

Figura 1. Microstructural development in a single stiff layer embedded in a soft matrix. The left and right columns represent, respectively, low and high driving force for film destabilization, and the top and bottom rows are for onset of instability and well after film break-up, respectively[Chirranjeevi et al. 2009].

2. Systems and Processing techniques

ATG instability is observed in a wide range of systems and processing techniques. Molecular Beam Epitaxy(MBE) is a technique where ATG is observed very frequently. In MBE, there is a solid substrate over which a thin film is grown. The mis-match in the lattice spacing of the substrate and the thin film can lead to the accumulation of elastic strain energy in the system. This kind of instability has been reported in SiGe/Si, GaInAsP/(001)InP systems. Hence, this is of great importance as these systems find their application in electronic and photonic device applications.

Self-assembly of Quantum dots can also be explained through ATG instability. Self-assembly refers to the spontaneous ordering of a dis-ordered structure into ordered structure. Quantum dots can be grown on substrate with pits so that there is minimal disorder in the system[Aqua and Xu 2015]. ATG instability in these systems is also referred to as Stranski-Krastanov growth.

The growth of thin films can be monitored by Auger electron spectroscopy (AES), reflection high energy electron diffraction (RHEED) and low-energy electron diffraction (LEED). In the above mentioned techniques the size of the probe may be greater than

the size of the islands. This may pose certain difficulties. To overcome these challenges, microscopic techniques like Transmission Electron Microscopy(TEM), Atom Force Microscopy (AFM), Scanning Tunneling Microscopy (SFM) etc can be used. These microscopes offer huge advantages as the magnification attainable is sufficient enough to visualize the islands.

3. Physical processes involved

In MBE, a thin film is grown over a substrate. Initially, the atoms tend to form atomically flat surface to reduce the surface energy. If there is a difference in lattice spacing between the film and the substrate, there will be a mis-fit strain in the film which will add to the elastic energy of the system. As the film grows, it becomes energetically favourable for the flat surface to break into "Isolated Islands". In this process, new surfaces are created which increases the surface energy but is compensated by the release of elastic energy.

4. Experimental parameters that control the instability

Temperature at which the process is carried out plays a significant role in determining the stability of the surface. The difference in the thermal expansion coefficients of the film and the substrate determines the mis-fit strains at various temperature. Temperature also determines the predominant mode of mass transport. The mechanism by which the atoms move from location to other determines the growth rate of the instability.
The thickness of the film influences the amount of strain in the film. Fig. 2 shows the XRD pattern of TiN thin films for various thicknesses. The amount of strain in the lattice can be calculated from the shift in the XRD peaks. The strain in the film determines the driving force for the instability.
The surface morphology of the substrate also plays an important role in the stability of the thin film. Pits on the surface of the substrate can lead to uniform distribution in the size of the islands formed[Aqua and Xu 2015].

5. Analysis of the interface

5.1. Governing Equations

The chemical potential of the particles at the surface is given in eq. 1.

$$\mu(x) = \mu_0 + \gamma\Omega\kappa(x) - \sigma_{nn}(x)\Omega, \tag{1}$$

Where, μ_0 is the chemical potential of the equilibrium flat interface bounding an unstressed solid. The second term in eq. 1 gives the contribution of the surface energy to the chemical potential.Where, γ is the interfacial tension, Ω is the atomic volume and κ is the curvature. The third term in eq. 1 is the contribution of the normal stress (σ_{nn}), to the chemical potential. However, in the present scenario the normal stress is zero as it is a free surface and hence the term is neglected.
Eq. 1 needs to be slightly modified to incorporate the effect of uniform stress in the material. Thus, the chemical potential of the system is given in eq. 2.

$$\mu(x) = \mu^* + \gamma\Omega\kappa(x) + \left([\sigma_{\tau\tau}(x)]^2 - \sigma^2\right)\frac{\Omega}{2M}, \tag{2}$$

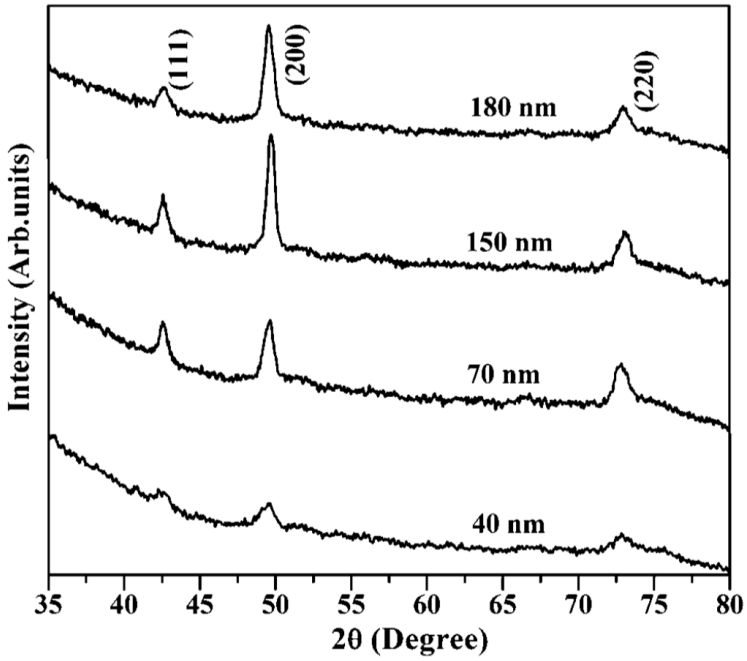

Figura 2. XRD pattern of TiN thin films for various thicknesses [Vasu et al. 2014].

Where, μ^* is the chemical potential of the flat surface with mean stress σ, M is the elastic modulus in the appropriate direction.

The phenomenon through which mass transport occurs determines the mode of diffusion. Mass transport can occur through surface diffusion, evaporation/condensation or through volume diffusion. In this present report mass transfer through surface diffusion is only considered.

For a surface diffusion controlled mass transport the atomic velocity along the surface and the chemical potential can be related through Nernst-Einstein relation given in eq. 3.

$$V = -\frac{D_s}{KT}\frac{\partial \mu}{\partial s}, \tag{3}$$

Where, D_s is the diffusivity along the surface and the derivative of chemical potential is taken with respect to the surface(s). This then could be related to the surface profile as given in eq. 4.

$$\frac{\partial h}{\partial t} = \frac{D_s \Omega \delta}{KT}\frac{\partial^2 \mu}{\partial s^2}, \tag{4}$$

Where, h is the surface profile of the interface and δ is the number of atoms per unit area.

Thus, the governing equation of the system is given by eq. 2 and eq. 4.

5.2. Linear stability analysis

Linear stability analysis (LSA) can be performed to understand the stability of the interface. LSA may not be able to predict the final equilibrium morphology but can successfully predict the stability of an interface, critical wavelength and the wavelength corresponding to the maximum growth rate.

Let us assume that $h = \Delta sin(kx)$ and $\Delta k << 1$. In the limit that the perturbations are small, the third term in eq. 2 can be simplified. In this present scenario the normal stress must be zero. Applying these constraints,

$$\left([\sigma_{\tau\tau}(x)]^2 - \sigma^2\right)\frac{\Omega}{2M} = \frac{2\Omega k\sigma^2}{M}\sin(kx), \tag{5}$$

Similarly, the curvature of the surface can be approximated as given in eq. 6 for small perturbations.

$$\kappa(x) = -h_{xx}(1 + {h_x}^2)^{-\frac{3}{2}} \approx -h_{xx} \tag{6}$$

Incorporating eq. 5 and eq. 6 into eq. 2 and then evaluating eq. 4, yields

$$\frac{\partial\Delta}{\partial t} = -C\gamma\Delta k^4 + \frac{2C\Delta\sigma^2k^3}{M}, \tag{7}$$

Where, $C = D_s\Omega^2\delta/KT$
Thus, the perturbation starts to grow for $k < k_c(\frac{2\sigma^2}{M\gamma})$ and the fastest growing wavenumber corresponds to $k_{max} = \frac{3}{4}k_c$.

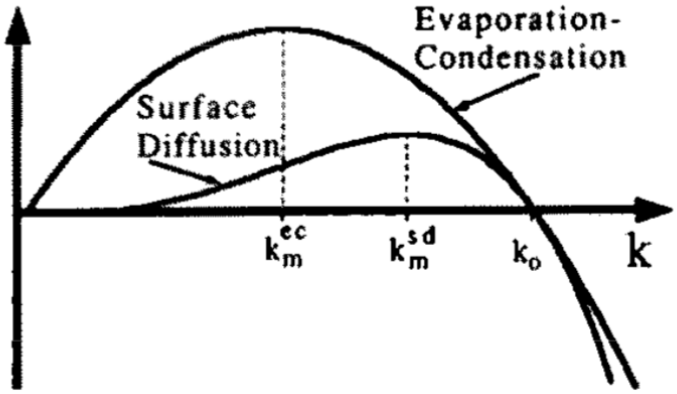

Figura 3. Growth rate for surface perturbations of wavenumber K. The two curves indicate different modes of mass transport[Srolovitz 1989].

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