Stress relaxation in thin films due to grain boundary diffusion and dislocation glide

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Chapter 7

Summary & Outlook

The aim of this study was to investigate inelastic deformation mechanisms in thin film–substrate systems in order to contribute to the understanding of size effects induced by the size of columnar grains. In this chapter the main conclusions from the previous chapters are summarized and an outlook for complete coupling between film growth, grain boundary diffusion and dislocation glide is given.

7.1 Summary

In Chapter 3 we have investigated the relaxation of intrinsic stresses by dislocation glide only. The intrinsic stress was represented by an array of grain boundary (GB) dislocations. The displacement discontinuity along the GBs is adjusted through the Burgers vector distribution to account for the realistic distribution of extra material that has been diffused into the GBs. Moreover, discrete dislocation (DD) simulations for film–substrate systems in the absence of intrinsic stress but instead loaded with uniform thermal stress were carried out. Interpretation of the results from these two types of calculations allowed to isolate the effects of initial stress distribution and magnitude on intrinsic stress relaxation. For relaxation of intrinsic stress, two effects of the grain size on the amount of relaxation were found to be counteracting. As the grain size decreases, plastic hardening in the film increases due to the confinement of dislocation motion in between impenetrable grain boundaries (GB): films with a finer microstructure are harder. In contrast, the initial intrinsic stress distribution becomes more homogeneous as the grain size decreases, thus promoting relaxation, as diffused material can be distributed among closely spaced GBs. Thus in terms of relaxation of intrinsic stresses induced by the presence of extra material along the GBs, smaller may be softer. The cross-over between these two size-dependence trends lies somewhere between $d = 0.5$ μm and $d = 0.25$ μm for the cases considered in Chapter 3.

In Chapter 4 we have studied GB diffusion itself with the motivation of developing a tool for a future unified diffusion–glide modelling. Therefore we have chosen to describe the diffusion process in terms of the 'climb' motion of discrete dislocations along the GBs. The governing equations were derived and a DD scheme was developed, followed by its application to the relaxation of thermal stress in stationary thin film–substrate system problems. Since this framework was two–dimensional, we were able to calculate, for the first time, the stress distribution inside the film during any instant of relaxation as well, rather than just the stress distribution along the GB. The main finding of Chapter 4 is that even when the steady state is attained for GB diffusion, a residual (unrelaxed) stress remains inside the film. This stress level depends on the aspect ratio of the grains (film thickness over grain width) and on the magnitude of the initial thermal stress. More specifically, our results revealed that the amount of relaxation was connected to the shape and width of the GB wedge. This shape resembles a U–like profile for fine grains and becomes a V–like profile for wide ones.

The DD model for GB diffusion highlighted the importance of the physical boundary conditions at the film–substrate interface. While existing diffusion models in the literature have incorporated the no-flux condition at the border, the condition that the GB wedge needs to vanish there has been ignored. Therefore, in Chapter 5 a two–dimensional continuum model was developed that does take all boundary conditions into account. The surplus of boundary conditions in the classical sense has been circumvented by means of a staggered numerical solution of diffusional and linear elastic boundary value problems, using a mix of finite difference and finite element procedures. This continuum framework is then adopted for the problem described in Chapter 4 where the thermal stress in a film–substrate system is relaxed by the transport of material from the free surface into the GBs. The calculations revealed a power law scaling between the efficiency of relaxation and the grain size, irrespective of the initial stress level. In contrast the amount of initial stress was found to be important in Chapter 4 when the DD framework was used. The comparison of DD and continuum predictions substantiated the diffusional size effects present in thin films, captured by the DD method only. The underlying reason is that while the GB wedge can take any value in the continuum model, in the DD model it is quantized by the Burgers vector (as a representation of the atomic size). Accordingly in the DD framework, the effectiveness of relaxation saturates at a grain aspect ratio when the GB wedge width is comparable with the the Burgers vector.

In Chapter 6 we have revisited the intrinsic stresses concept, this time by looking into the development of the compressive intrinsic stress as a consequence of the supersaturation of surface adatoms during deposition. The DD and continuum models were extended for growing films with the aim of studying parameters that govern the film stress as a function of the growing film thickness. Two key process/material parameters have been identified. In accordance with our previous findings, the smaller the columnar grains, the more effective is GB diffusion; therefore the initial tensile coalescence stress is rapidly relaxed and subsequently the compressive deposition stress dominates. The second important controlling parameter is the growth rate of the film. When the growth rate is low or the temperature is high (the effective diffusion coefficient is exponentially dependent on temperature), diffusion is more effective. By contrast, in case of fast growth or low temperature, GB diffusion enforces a compressive stress lower in magnitude. For the extreme cases where both film growth is fast and film grains are wide, a fraction of the initial tensile coalescence stress persists.

7.2 Outlook

The coupling of GB diffusion and dislocation glide for a growing film can be achieved by incorporation of some ingredients from Chapter 3 to the model presented in Chapter 6. The first step is to introduce slip planes as described in Chapter 3. Frank–Read sources placed randomly on these slip planes generate dislocation dipoles that glide according to the Peach–Koehler force and drag controlled mobility. The mobility of glide dislocations is generally much higher than that of the GB dislocations representing GB diffusion at the temperature of interest. Therefore the timescales of the two relaxation mechanisms are uncoupled. This timescale separation can be exploited in the model by calculating the dislocation evolution by solely considering glide dislocations until all of them are either stopped at the impenetrable boundaries or got entangled with dislocations on other slip planes or exit from the free surface. Once this happens the GB diffusion simulation is re-activated. However, if any of the Frank–Read sources gets triggered during this simulation due to the changing stress state, the procedure is switched back to the short-time dislocation glide scheme. Simultaneously film is growing with a constant deposition rate. In Chapter 6 it has been shown that the time scale for growth relative to GB diffusion can be addressed by the normalized growth rate $\dot{H}$. For the $\dot{H}$ values considered in Chapter 6 the time scale for film growth is also much greater than the characteristic time scale for dislocation glide. Hence film growth is also deactivated during any short-time dislocation glide scheme.

The initial film thickness h_0 is so small that plasticity cannot be mediated by dislocation glide. Hence in the model initially no Frank–Read sources are present. In fact, Frank–Read sources are considered to exist only once the film has reached a critical thickness h_{cr} large enough that the average nucleation distance L_{nuc} (see Chapter 2) can be accommodated on slip planes. Until this thickness is achieved, dislocation glide is disabled and inelastic deformation is due to GB diffusion only, just like in Chapter 6.

For simplicity we assume that the film has a constant density of sources, ρ_{nuc}, randomly sprinkled over slip planes that have an orientation ϕ relative to the film–substrate interface. When the film thickness exceeds h_{cr}, Frank–Read sources are added in order to maintain the constant ρ_{nuc} in accordance with the total area of film material. When a source is to be added, a random slip plane is selected and subsequently the source is positioned at a random location on that slip plane (yet at a safe distance of L_{nuc} away from GBs or the film–substrate interface). Nucleation of dislocation dipoles from sources is governed by the criterion described in Chapter 3: that is, if the resolved shear stress on the source position exceeds the source strength τ_{nuc} for a sufficiently long timespan t_{nuc}, a dislocation dipole with separation L_{nuc} is introduced. During growth slip planes are extended together with film thickness.

For an initial trial we have considered an infinitely wide film with a grain size of $d = 0.25\,\mu$m having two slip systems with orientations $\phi = 60°$and $\phi = 120°$. Potentially active slip planes in the film are taken to be separated at $200b$. The initial film thickness is $h_0 = 0.02\,\mu$m and it increases according to a normalized growth rate $\dot{H} = 54$. At this initial stage a uniform tensile coalescence stress $\sigma_0 = 250\mathrm{MPa}$ is assumed to exist in the film and the value of the compressive stress enforced by the adatom supersaturation at the free surface is $\sigma_s = -250\mathrm{MPa}$. The critical thickness $h_{\mathrm{cr}} = 0.1\,\mu$m is decided on the basis of the nucleation distance L_{nuc} whose average value $L_{\mathrm{nuc}} = 0.0625\,\mu$m follows from the mean nucleation strength $|\tau_{\mathrm{nuc}}| = 25\mathrm{MPa}$. Until $h = h_{\mathrm{cr}}$ dislocation glide is

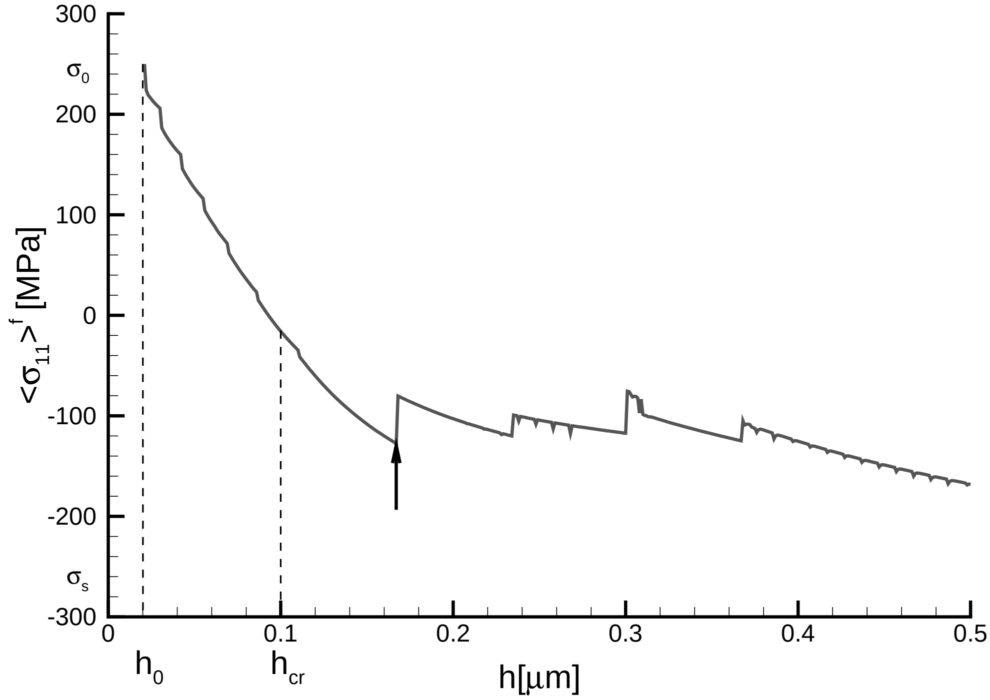

Figure 7.1: Film average stress evolution for a growing film, taking into account GB diffusion and dislocation glide.

disabled, and only GB diffusion takes place by way of the nucleation and climb of GB dislocations.

Experimental investigations of in-situ stress evolution during Volmer–Weber type growth of thin films on a substrate have revealed the typical graph shown in figure 3.1 of the film average stress vs. film thickness (see Chapter 3). The essence of that graph is that the peak tensile stress is reached once full coalescence is attained, and that further growth leads to relaxation and the development of compressive stress. The simulated development of the film average stress given in figure 7.1 exhibits these features. For $h < h_{\mathrm{cr}}$, diffusional relaxation has just relaxed the coalescence stress and a small amount of compressive stress in film average sense has emerged, just like in Chapter 6. When the critical thickness is reached, Frank-Read sources are started to be added to the film. The number of sources is calculated from the dislocation density which is chosen to be $\rho_{\mathrm{disl}} = 60\,\mu\mathrm{m}^{-2}$. However, the activation of sources depends on the resolved shear stress at the source positions and in this example the first glide dislocations were generated at a thickness of around $h = 0.17\ \mu$m (marked by an arrow in figure 7.1). At the moment, stress relaxation by dislocation glide commences. Since this happens on a timescale that is much shorter than that of growth, the stress development due to glide emerges as a stress jump in figure 7.1. Once these initial dislocations get halted at the GB or leave the crystal through the free surface, diffusion and film growth continue. Up to a thickness of

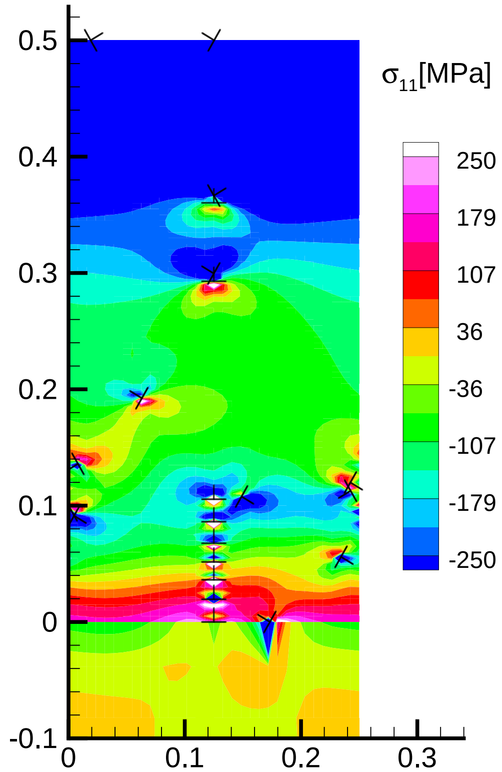

Figure 7.2: Stress distribution and dislocation positions when $h = 0.5\ \mu$m

$h = 0.5\ \mu$m, four major bursts of dislocation nucleation and glide occur in this example, yet interspersed with numerous short periods of plasticity.

The stress distribution together with the dislocations in the final state, given in figure 7.2, suggests that glide dislocations that got pinned at the GB may inhibit further diffusion. Depending on the orientation of the slip planes, the stress field of pinned glide dislocations may have a compressive 11-component that would repel the GB dislocations traveling towards that point from the free surface. Evidently, the result presented above is just an example for a specific set of parameters. In order to disclose the predisposition of growing film–substrate systems to relaxation a systematic study should be performed to asses role of the variables $\rho_{\rm nuc}$, $\tau_{\rm nuc}$, d, $\dot{H}$ independently. Also multiple realizations will be needed to average over different dislocation source structures.