MULTI-SCALE MODELING OF DEFORMATION IN POLYCRYSTALLINE THIN METAL FILMS

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ABSTRACT

The time-dependent irreversible deformation of thin metal films on substrates is investigated on various levels of modeling. The work focuses on polycrystalline films without cap layer, where diffusive processes play an important role. A continuum level model of constrained grain boundary diffusion describes how surface and grain boundary diffusion lead to the relaxation of tractions along the grain boundaries. If the film is perfectly bonded to the substrate, the relaxation of tractions along the grain boundary leads to a crack-like stress concentration close to the film/substrate interface. By virtue of this stress concentration, dislocations on glide planes parallel to the thin film are subject to a driving force that does not exist in the case of purely bi-axial loading. This model gives a theoretical basis for experimental observations of such parallel glide processes.

To obtain a better understanding of the microscopic phenomena associated with the this creep process, we conducted large scale molecular dynamics (MD) simulations, as well as discrete dislocation dynamics (DDD) studies on intermediate length scales. The results of these numerical simulations reveal that nucleation of glissile dislocations at the tip of the diffusion wedge can be described with a critical stress intensity factor criterion. The validation of this criterion for dislocation nucleation by MD simulations allows transferring this information onto the DDD length and time scales. The deformation behavior of thin films is investigated with DDD simulations on length scales from nanometers to microns. If only dislocation glide is active, DDD simulations with a constant density of dislocation sources show that the flow stress of thin films scales linearly with the inverse film thickness. Furthermore, the DDD simulations reveal that, upon activation of the creep mechanisms in ultra-thin films the size scaling of the flow stress breaks down. Instead, such films show a thickness-independent flow stress. These findings agree well with experimental observations.

1 INTRODUCTION

The understanding of deformation mechanisms in small scale structures is one of the outstanding fundamental problems in materials science to be solved. The advent of nanostructured materials and the need to design microelectronic and micromechanical devices with typical length scales on the order of nanometers requires not only a profound understanding, but also a reliable prediction of mechanical properties of such structures. Experiment shows that the flow stress in the micrometer regime increases inversely proportional to the film thickness (see for example Keller *et al.* [1]). This has been attributed to dislocation channeling through the thin film by Nix [2], where a threading dislocation moves through the film, leaving behind interfacial dislocation segments. The energetical effort to generate these interfacial dislocations increases with decreasing film thickness. This model, however, could not completely explain the high strength of thin films found in experiments as discussed by Keller *et al.* [1]. More recent theoretical and experimental work by Legros *et al.* [3], von Blankenhagen *et al.* [4] and Balk *et al.* [5] indicates that the strength of thin metal films results from a lack of dislocation sources rather than from the energetic effort associated with dislocation motion. For yet thinner films – with thicknesses below 500 nm – Balk *et al.* [5] showed that the flow stress becomes independent of the film thickness. This transition is accompanied by the occurrence of dislocations gliding on planes parallel to the film/substrate interface. This indicates that large, long-range internal stresses are built up that decay only slowly on the length scale of the film thickness, because in the global equibiaxial stress there is no resolved shear stress on such glide planes.

A possible source for these internal stresses has been identified in the directional diffusion in thin films constrained by a substrate. In these structures atoms migrate from the free top surface into the grain boundaries. The directed diffusion in the columnar microstructure of thin films on substrates thus relaxes the tractions along the grain boundaries from the top to the root. Due to the perfect bonding of the film to the substrate, a stress concentration at the root of the grain boundary builds up, leading to a singular, crack-like stress field in the film as the grain boundary tractions are relaxed. Because the material inserted into the grain boundary by this one-directional diffusion takes the shape of a wedge, this defect has been referred to as diffusion wedge by Gao *et al.* [6]. The formation of diffusion wedges by grain boundary diffusion and the subsequent nucleation of parallel glide dislocations has been investigated with molecular dynamics (MD) simulations by Buehler *et al.* [7, 8], which confirmed that the displacement field near a diffusion wedge indeed becomes crack-like. The crack-like stress field allows defining a stress intensity factor that characterizes the deformation field. Furthermore, it was shown that the nucleation of parallel glide dislocations follows a critical stress intensity factor criterion. Based on this atomistic information, a mesoscopic dislocation-based model for deformation in thin films has been developed. First applications of this model in Buehler *et al.* [8] showed that that the creep mechanism of grain boundary diffusion coupled with the subsequent nucleation of parallel glide dislocations can indeed cause a constant flow stress.

In this work we investigate the transition between the deformation regimes with conventional dislocation slip and diffusional creep with a discrete dislocation dynamics (DDD) method, where inelastic deformation is described as climb and glide of dislocations. In the following we present the DDD model describing dislocation plasticity in the geometry of a thin film on a substrate. The application of this model allows us to calculate macroscopic quantities, like the flow stress that can immediately be compared with experimental results. By this comparison we can conclude on the limiting mechanisms of plasticity in thin and ultra-thin films.

2 MODEL

The model used in this work follows the well-known discrete dislocation models in two and three dimensions described in the literature (see for example [4, 9, 10] for thin film plasticity). In such models, dislocations are considered sources of stress and strain in a linear elastic continuum. In the two-dimensional model underlying this work, the stress and strain fields of straight dislocations in a bi-crystal are calculated according to Mura's solution [11]. This solution allows films on substrates with different elastic properties to be studied. However, in the present work, film and substrate have identical elastic properties. The free surface of the film is established by the decomposition of the problem into calculating stress and strain in an infinite medium and compensating the surface tractions by counter forces. These counter forces are stemming from a surface layer of virtual dislocations with non-lattice Burgers vectors. The distribution of these virtual dislocations that compensates the surface tractions is calculated with a boundary integral method. The Green's function for the boundary integral is constructed following the approach of Gutkin and Romanov [12]. Once the surface distribution of the virtual dislocations is known, their stress and strain field within the medium can again be calculated by evaluating Mura's equations. The details of this method are described elsewhere [13]. No special boundary conditions are employed at the lateral boundaries of the simulation domain, rendering the geometry effectively to an infinite strip of material on a substrate. However, dislocations that pass the lateral perimeter of the simulation box or its top surface are excluded from the simulation. Dislocations the hit the film/substrate interface are immobilized, rendering this interface into an impenetrable barrier for dislocations. As indicated in Figure 1, three slip planes are considered in the work. Two are inclined by 45° with respect to the plane of the thin film, and one slip plane originating from the root of the grain boundary is parallel to the film/substrate interface to incorporate parallel glide dislocations into the DDD simulation.

With this simulation scheme, the elastic interactions of all climb and glide dislocations contained within a thin film structure can be calculated accurately. Once the elastic driving force on a dislocation is known, its velocity can be calculated. In the following, we assume overdamped dislocation motion, *i.e.* the dislocation velocity is directly proportional to the driving force. Dislocation climb is assumed to occur 100 times slower than dislocation glide under the same driving force. The lateral dimension of the simulation domain that defines the grain size-film thickness ratio is kept at a constant value of $L = 3h_f$ throughout this work.

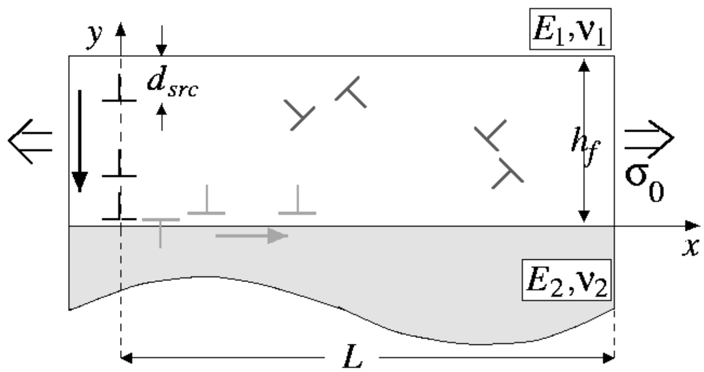

Figure 1: Sketch of the considered two-dimensional slip geometry in a thin film on a substrate. Climb dislocations are drawn in black, parallel glide dislocations in light gray and dislocations on inclined slip planes in dark gray.

The phenomenological rules for dislocation nucleation are explained in the following. As shown in Figure 1 two essentially different kinds of dislocation sources are considered. The rule determining generation of a new climb dislocation is based on the force on a test dislocation at a given source position in distance d_{src} from the surface. If the total force drives this test dislocation into the film, the dislocation is considered as being nucleated and it further participates in the dynamical simulation in the usual manner. This method gives an upper limit for the dislocation generation rate, because all dislocations that can self-consistently be nucleated from a fixed source are effectively produced. These dislocations climb along the grain boundary and pile-up against the film/substrate interface, which leads to a crack-like stress concentration. Thereby the climb pile-up effectively represents a grain boundary diffusion wedge.

The criterion for generation of glide dislocations follows similar lines, except that a dislocation dipole is used for testing rather than a single dislocation, representing the two dimensional analogon of a Frank-Read source. The critical stress to separate the dislocations in the dipole is then dependent on the dipole width as well as on the local shear stress. A new dislocation dipole in created if and only if the local shear stress does at least hold the dipole in equilibrium. If the dipole collapsed under the local shear stress, self-consistent dislocation nucleation from this source would not possible and is consequently not considered in the simulation. This method consequently gives again an upper limit for the production rate of glide dislocations. The phenomenological rules for the nucleation of climb and glide dislocations critically depend on input parameters. The critical parameter for the nucleation of climb dislocations is the distance of the dislocation source to the surface. In the present work this parameter is set to a constant value according to the findings in [13]. The dipole separation for all glide dislocation sources is chosen such that the dipoles are stable under a shear stress of $\tau_{crit} = 0.6$ GPa. To avoid all sources becoming active at the same time, the dipole separations are randomly varied to give a normal distribution with a variance of 10 % of the mean value. For the nucleation of *parallel* glide dislocations at the root of the grain boundary the distance of the dislocation source to the diffusion wedge also determines the critical local stress. The position of the dislocation source from the diffusion wedge is adjusted such that the critical level for dislocation nucleation is reached, when the effective stress intensity factor of the diffusion wedge reaches the critical value obtained from MD simulations [14]. Thus the MD simulations are used to validate critical parameters of the DDD scheme in a hierarchical multiscale simulation approach. Further rules have to be applied to handle close encounters of two dislocations. If two dislocations of opposite Burgers vector approach each other closer than 6 Burgers vectors, they annihilate and are removed from the simulation. If two dislocations with the same or non-parallel Burgers vectors approach closer than that distance they are immobilized and form a lock.

3 RESULTS

In this work, the flow stress in a thin film is calculated by straining the film elastically up to a stress of 2 GPa and then allowing it to relax plastically. After a given time the stress remains at a constant level, defining the flow stress of the film. Three deformation modes are considered: (1) deformation by grain boundary diffusion and parallel dislocation glide, (2) deformation with dislocation glide on inclined slip planes, and (3) deformation with all deformation mechanisms being active. The creep relaxation mode 1 has been studied in detail in previous work [13], where it has been shown that the creep relaxation gives a constant flow stress if a fixed position d_{src} of the dislocation source for climb dislocations is assumed. Here we focus on the plastic relaxation under dislocation slip on inclined slip planes (mode 2) and the combination of both relaxation modes (mode 3).

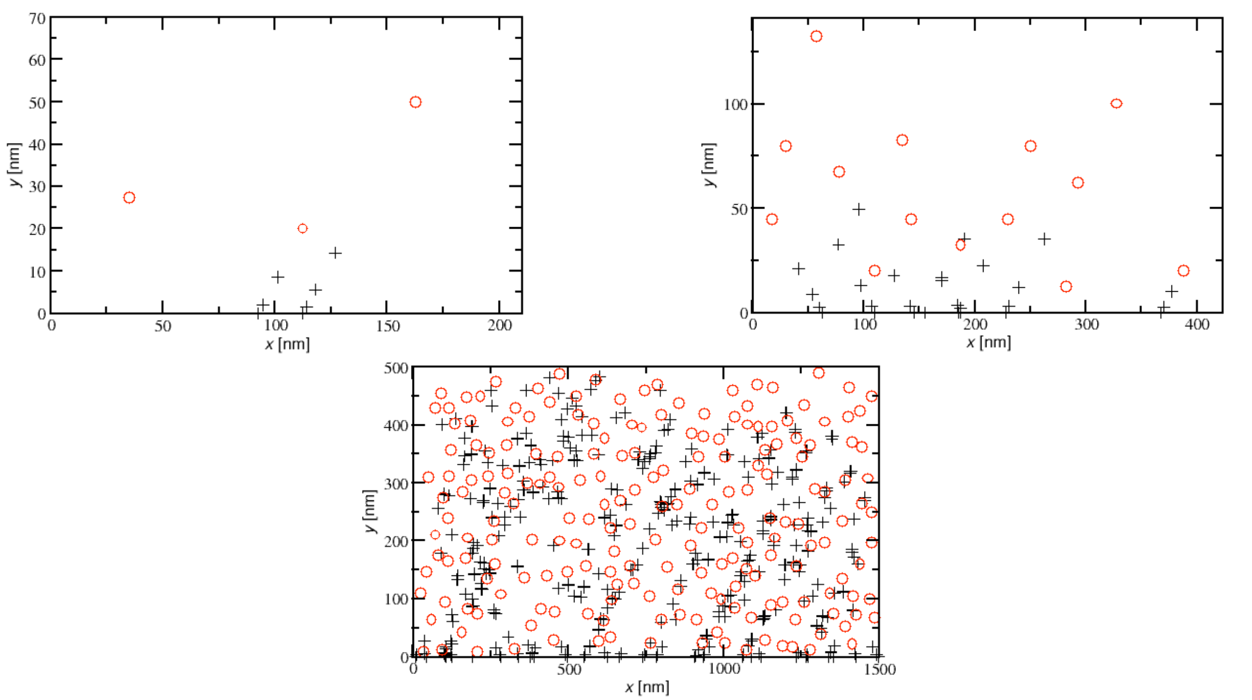

Figure 2: Final dislocation configurations for films of thickness 70nm (top left), 140nm (top right), and 500nm (bottom) relaxed under dislocation slip on inclined planes. Circles indicate the positions of dislocation sources, crosses show the positions of the dislocations.

The final dislocation configurations for films of different thicknesses that have been relaxed under deformation mode 2 are shown in Figure 2. It is seen that the dislocations pile-up against the film/substrate interface until their back-stress effectively shuts down further dislocation nucleation at the sources. We also note that the dislocation distribution in the thickest film investigated here (film thickness 500nm) is quite homogeneous in the middle of the film with only a small decrease close to the surface and a small increase towards the substrate.

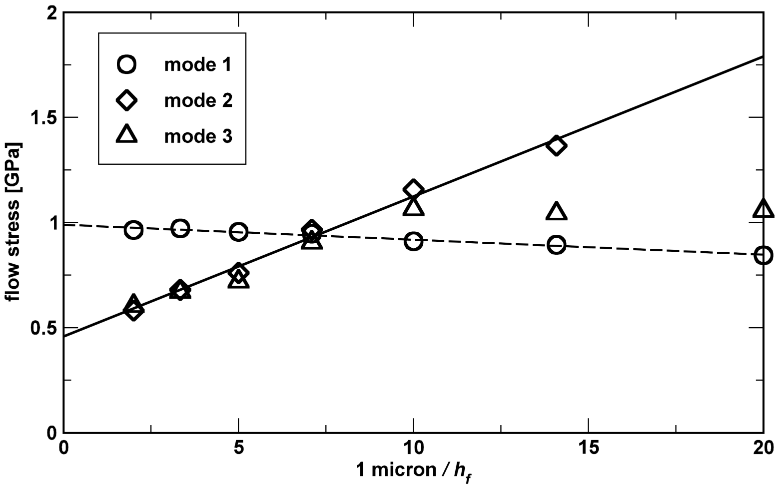

Figure 3: Flow stress versus inverse film thickness for different deformation mechanisms as given in the legend

The resulting flow stress for all three deformation modes is plotted in Figure 3. The results obtained for dislocation slip on inclined planes exhibit a linear increase in flow stress with inverse film thickness. The source density of sources for inclined dislocation slip amounted to 130 sources / μm^3. It is seen that for these parameters strain relaxation by dislocation slip is more effective than the creep mechanism for thicker films. At a film thickness of 140 nm a transition occurs and the diffusion mechanism can reduce the stress in thinner films film to lower values. The simulation with creep and slip on inclined slip planes shows that the deformation mechanisms cannot be considered independent of each other. In the slip dominated regime diffusional creep has no influence on the results. However, the dislocations on inclined slip planes have the potential to block dislocation climb if they intersect the grain boundary. Thus, the flow stress in the creep dominated regime is slightly higher in mode 3 than for pure creep deformation and parallel slip (mode 1). The flow stress values for the deformation modes 1 and 2 can be interpolated very well by straight lines with a positive offset at the stress axis. For the dislocation glide mode this stress offset is close to the value of the activation stress of the dislocation sources, which represents the yield stress of the model material.

4 DISCUSSION

The two-dimensional discrete dislocation dynamics study of plasticity in thin films shows that the different deformation mechanisms proposed for different film thicknesses can account for the experimentally found transition between the regime of flow stresses depending inversely on the film thickness and the thickness independent regime for ultra-thin films. The simulations revealed that diffusional creep is an effective mechanism of plastic relaxation for ultra-thin films. This creep mechanism yields essentially a constant flow stress independent of the film thickness. Dislocation slip on inclined slip planes, as alternative deformation mechanism, is restricted by the back stress of dislocation pile-ups at the film/substrate interface. Therefore the flow stress obtained by this mechanism shows a linear dependence on the inverse film thickness. The combination of both deformation mechanisms consequently reveals that the inclined slip mechanism will be dominant in thicker films, while the creep mechanism prevails in ultra-thin films. It is also seen that dislocations on inclined slip planes interfere with parallel glide dislocations and thus effectively shut down the creep mechanism, which results in slightly higher values for the flow stress than for pure creep deformation.

A key ingredient in obtaining a flow stress inversely proportional to the film thickness was that the source density for all film thicknesses is constant. In other work on dislocation modeling of thin film plasticity [10] such strict size scaling could not be observed. The reason for the increasing flow stress in geometrical confinement lies in the back stress of the dislocations piling up against the film substrate interface. This back-stress effectively stops further dislocation nucleation at the dislocation sources. *In-situ* experiments under transmission electron microscope (TEM) observation [5] do not reveal dislocation pile-ups or even multiple activation of Frank-Read-like dislocation sources. Instead, dislocations are seen to originate mainly from grain boundaries. In this respect we conclude that the model employed in this work gives an upper limit for plastic relaxation with inclined slip, because the sources are evenly distributed within the volume and can operate multiple times. Even under these conditions, beneficial for plasticity, the geometrical confinement has a pronounced influence on the flow stress. However, future work should make use of refined dislocation generation mechanisms adapted to experimental observations.

5 REFERENCES

[1] Keller, R.-M., Baker, S.P., and Arzt, E., Quantitative analysis of strengthening mechanisms in thin Cu films: Effects of film thickness, grain size, and passivation, J. Mat. Res. **13**, 1307-1317, 1998.

[2] Nix, W.D., Yielding and strain hardening of thin metal films on substrates, Scripta mater. **39**, 545-554, 2001.

[3] Legros, M., Hemker, K.J., Gouldstone, A., Suresh, S., Keller-Flaig, R.-M., and Arzt, E., Microstructural evolution in passivated Al films on Si substrates during thermal cycling, Acta mater. **50,** 3435-3452, 2002.

[4] Blanckenhagen, B. von, Gumbsch, P., and Arzt, E., Dislocation sources and the flow stress of polycrystalline thin metal films, Phil. Mag. Lett. **83**, 1-8, 2003.

[5] Balk T.J., Dehm, G., and Arzt, E., Parallel glide: unexpected dislocation motion parallel to the substrate in ultrathin copper films, Acta mater. **51**, 4471—4485, 2003.

[6] Gao, H., Zhang, L., Nix, W.D., Thompson, C.V., and Arzt, E., Crack-like grain-boundary diffusion wedges in thin metal films, Acta mater. **47**, 2865—2878, 1999.

[7] Buehler, M.J., Hartmaier, A., and Gao, H., Atomistic and continuum studies of crack-like diffusion wedges and associated dislocation mechanisms in thin films on substrates, J. Mech. Phys. Sol. **51**, 2105-2125, 2003.

[8] Buehler, M.J., Hartmaier, A., and Gao, H., Hierarchical multi-scale modeling of plasticity of submicron thin metal films, Mod. Sim. Mat. Sci. Engrg. , 2003.

[9] Hartmaier, A., Fivel, M.C., Canova, G.R., and Gumbsch, P., Image stresses in a free-standing thin film, Mod. Sim. Mat. Sci. Engrg. **7**, 781-793, 1999.

[10] Nicola, L., Van der Giessen, E., and Needleman, A., Discrete dislocation analysis of size effects in thin films, J. Appl. Phys. **93**, 5920—5928, 2003.

[11] Mura, T., The Continuum Theory of Dislocations, in Advances in Materials Research, Vol. 3, Ed. H. Herman, Interscience Publishers, New York, 1968.

[12] Gutkin, M.Y., and Romanov, A.E., Straight edge dislocations in a thin two-phase plate, physica status solidi **125**, 107-125, 1991.

[13] Hartmaier, A., Buehler, M.J., and Gao, H., A discrete dislocation plasticity model of creep in polycrystalline thin films, Defects and Diffusion in Metals - An Annual Retrospective VI - Defect and Diffusion Forum, Vol. 224-225, pp. 107-125, 2004.