ADVANCED
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Multiscale Modeling of Deformation in Polycrystalline Thin Metal Films on Substrates

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The time-dependent irreversible deformation of a thin metal film constrained by a substrate is investigated by a mesoscopic discrete dislocation simulation scheme incorporating information from atomistic studies of dislocation nucleation mechanisms. The simulations take into account dislocation climb along the grain boundaries in the film as well as dislocation glide along slip planes inclined and parallel to the film/substrate interface. The calculated flow stress and other features are compared with relevant experimental observations. The work is focused on deformation of a polycrystalline film without a cap layer, for which diffusive processes play an important role. The dislocation-based simulations reveal information on the prevailing deformation mechanisms under different conditions and for different film thicknesses. Despite of the limitations of the two-dimensional dislocation model, the simulations exhibit a film thickness dependent transition between creep dominated and dislocation glide dominated deformation, which is in good agreement with experimental observations.

1. Introduction

Miniaturization of structures and devices in micro- and nanotechnology leads to the development of materials and compounds with novel properties that cannot simply be extrapolated from material properties on larger scales. Mechanical behavior and reliability of devices containing thin metal films are of critical importance to innovations in integrated microelectronic, electro-mechanical, optoelectronic, and micro- or nano-electro-mechanical devices. In addition, biotechnology applications such as bio-chips and lab-on-a-chip devices are possible future technologies where ultrathin material layers with thicknesses well below 100 nm will be heavily used. In many applications and during the manufacturing process, thin films are subject to thermal stresses that are known to have a significant influence on performance and reliability of devices. Thus the understanding and the quantitative description of deformation mechanisms in small-scale structures is of paramount importance for the knowledge-based design of new materials and structures.[1]

Experimental observations and numerical simulations continue to improve our knowledge about deformation mechanisms in polycrystalline thin films. For example, it is known that the intragrain dislocation activity and grain boundary mediated diffusional creep are important mechanisms of inelastic deformation in these films.[2,3] Experiments on films with thicknesses in the submicrometer regime often reveal a flow stress inversely proportional to the film thickness.[1,4] For single crystalline films, this has been shown to be a result of dislocation channeling through the thin film,[5,6] where mobile dislocation segments threading the film thickness cause dislocation segments to be deposited at the film/substrate interface. Balancing the elastic energy released from the threading dislocation motion with that stored in the newly created length of interfacial dislocation yields a critical stress inversely proportional to the film thickness. Below this critical stress level, motion of threading dislocations is not possible. Recent experimental and theoretical work [2,6–8] indicates that the strength of polycrystalline thin metal films, which is usually 3 to 4 times higher than that of single crystalline films,[6] results from the difficulty to operate dislocation sources in the geometrical confinement of a thin film, rather than from the energetic effort associated with dislocation motion as for single crystalline films.

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A further interesting observation is that the flow stress becomes independent of the film thickness once it is below 500 nm.[2] This transition is accompanied by the occurrence of dislocations gliding on planes parallel to the film/substrate interface. Since there is no resolved shear stress on such glide planes, this indicates the presence of large internal stresses that decay quite slowly on the length scale of the film thickness. A possible source for these internal stresses has been identified in the directional diffusion in an uncapped thin film constrained by a substrate. In this process, atoms migrate from the free surface of the film into the grain boundaries. The directed diffusion in the columnar microstructure of a thin film on a substrate creates a defect referred to as diffusion wedge[9] which induces a crack-like stress field by relaxing the tractions across a grain boundary. Molecular dynamics (MD) simulations[10,11] confirmed that the displacement field near a diffusion wedge indeed becomes crack-like and can be characterized by a stress intensity factor. It was further shown that the nucleation of parallel glide dislocations follows a critical stress intensity factor criterion.[10] Based on this information from atomistic modeling, a mesoscopic dislocation-based model has been developed for creep deformation in thin films. First applications of this model showed that grain boundary diffusion coupled with subsequent nucleation of parallel glide dislocations can indeed cause a constant flow stress.[11,12]

In this paper, we briefly re-iterate the main findings from the two-dimensional discrete dislocation model and the MD simulations of diffusional creep in a polycrystalline thin film on substrate. The dislocation model is then generalized to include conventional dislocation glide on inclined slip planes in the film. This allows us to study how the deformation mechanism changes with reduced film thickness. The results are critically examined to reveal the limitations of two-dimensional dislocation models of thin film plasticity.

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 Refs [8,12–15] for thin film plasticity). In such models, dislocations are considered sources of stress and strain in a linear elastic continuum. The free surface of the film is established by a boundary integral method as described elsewhere.[12] No special boundary conditions are employed at the lateral boundaries of the simulation domain, rendering the geometry effectively into an infinite strip of material on a substrate. Dislocations that pass the lateral perimeter of the simulation box or its top surface are excluded from the simulation, while dislocations that hit the film/substrate interface are immobilized, effectively making this interface an impenetrable barrier. As indicated in Figure 1, taken into consideration are two slip systems inclined by ± 45 ° with respect to the plane of the film and a single slip plane parallel to the film/substrate interface originating from the root of the grain boundary.

With this simulation scheme, the elastic interactions among all climb and glide dislocations contained within the thin film can be calculated. Once the elastic driving force on a given dislocation is known, its velocity can be determined. We assume overdamped dislocation motion so that the dislocation velocity is directly proportional to the driving force. Dislocation climb is assumed to be 100 times slower than dislocation glide under the same driving force.[12] The lateral dimension of the simulation domain that defines the grain size-film thickness ratio is kept at a constant value of $L=3\ h$.

The phenomenological rules for dislocation nucleation are as follows. 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 subsequently participates in the dynamical simulation. This method gives an upper limit for the dislocation generation rate, as 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, leading to a crack-like stress concentration. The climb pile-up effectively represents a grain boundary diffusion wedge. 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 an earlier study.[12]

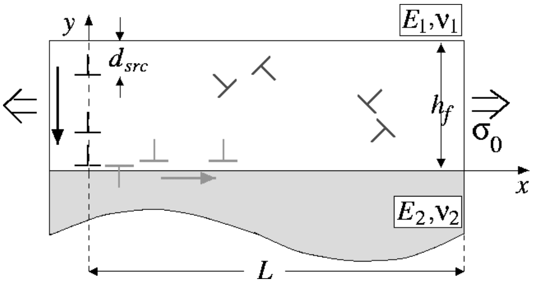

Fig. 1. Sketch of the considered two-dimensional slip geometry in a thin film on substrate. Climb dislocations are drawn in black, parallel glide dislocations in light grey and dislocations on inclined slip planes in dark grey.

The criterion for generation of glide dislocations follows similar lines, except that a dislocation dipole, rather than a single dislocation, is used for testing, representing the two dimensional analogue of a Frank-Read source. The critical stress to separate the dislocations in the dipole then depends on the dipole width as well as on the local shear stress. A new dislocation dipole is created if and only if the local shear stress is large enough to at least hold the dipole in equilibrium. If the dipole collapses under the local shear stress, self-consistent dislocation nucleation from this source under the given stress is not possible. This method consequently gives again an upper limit for the production rate of glide dislocations. The dipole separation for all glide dislocation sources is chosen such that the dipoles become stable under a shear stress of σ_{crit}=0.6 GPa, corresponding to the nominal (bulk) flow stress of the material.

For the nucleation of *parallel* glide dislocations at the root of the grain boundary, a critical parameter is the distance of the dislocation source to the diffusion wedge. The position of the dislocation source with respect to the diffusion wedge is adjusted such that dislocation nucleation occurs when the effective stress intensity factor of the diffusion wedge reaches the critical value obtained from MD simulations.[10] In this way, the MD simulations are used to validate critical parameters of the DDD scheme in a hierarchical multiscale simulation approach. The lattice distortions near the root of the grain boundary from atomistic and dislocation simulations immediately before the first parallel glide dislocation nucleates are shown in Figure 2.

Further rules are applied to handle close encounters of two dislocations. If two dislocations of opposite Burgers vector approach each other closer than 6 Burgers vectors, they are assumed to annihilate each other and are removed from the simulation. If two dislocations with identical or non-parallel Burgers vectors approach closer than that distance, they are assumed to form a lock and are subsequently immobilized.

3. Results

In this work, the flow stress in a thin film is calculated by straining the film elastically up to an initial stress and then allowing it to relax plastically. The plastic relaxation here is calculated based on the nucleation and motion of individual dislocations as described above. 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,[12] where it has been shown that the creep relaxation gives a constant flow stress if the dislocation source for climb dislocations is located at a fixed distance d_{src} from the free surface. In the present work, 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 3 shows the dependence of the flow stress on the initial stress (or initial elastic strain) for different film thicknesses. It is seen that the flow stress can be fitted with a linear equation if the film thickness is larger than 70 nm. The slope of the linear fit lines increases with the initial strain. The extrapolation of the fit lines towards infinitely thick films exhibits different axis offsets for different initial strain levels. In particular, the simulations with the smallest initial strain show that the flow stress found in these relaxation simulations can drop below the critical stress to nucleate dislocations, because the motion of dislocations nucleated just above the threshold stress causes considerable further plastic strain relaxation. Films thinner than 70 nm exhibit saturation of flow stress at values close to the initial stress level. For all these simulations, a constant density of 130 dislocation sources perμm^3 has been used. The differences in the flow stress stem from the back stress of dislocation pile-ups against the film/substrate inter-

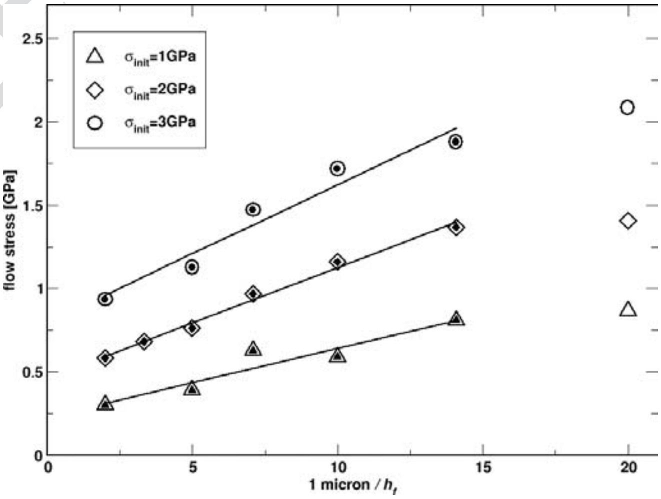

Fig. 3. Flow stress after relaxation in Mode 2 versus inverse film thicknesses for different initial stresses as given in the key. The data for larger film thicknesses (small filled symbols) is fitted with a linear equation, showing that slope increases with initial strain.

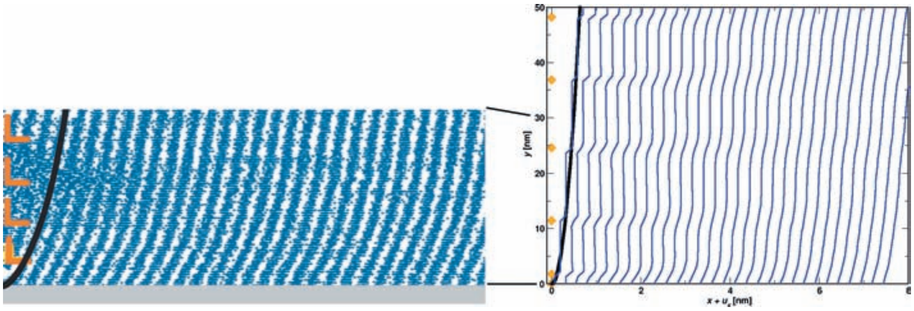

Fig. 2. Left: Distortion of the atomic lattice at the root of the grain boundary according to MD simulations[10]. The positions of effective dislocations in the grain boundary are marked. Right: Positions of climb dislocations in the pile-up (filled symbols) and resulting displacement field u_x (thin lines) as obtained by dislocation dynamics simulations. In both sub-figures, the elastic solution for the opening displacement of a crack tip with stress intensity factor 16 MPam$^{1/2}$ is plotted for comparison (thick line).

face that eventually shut down dislocation generation from a source. Thus, dislocation sources in smaller films produce fewer dislocations and consequently stop the plastic relaxation at higher stress levels.

The resulting flow stress for all three deformation modes is plotted in Figure 4 for a constant initial stress of 2 GPa. For thicker films with the employed simulation parameters, strain relaxation by dislocation slip on inclined slip planes is more effective than that by diffusional creep . A transition occurs at a film thickness of 140 nm, below which the diffusion mechanism becomes more effective, driving the stress to lower values. The simulation with combined 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, 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 in Mode 1 for pure creep deformation and parallel slip.

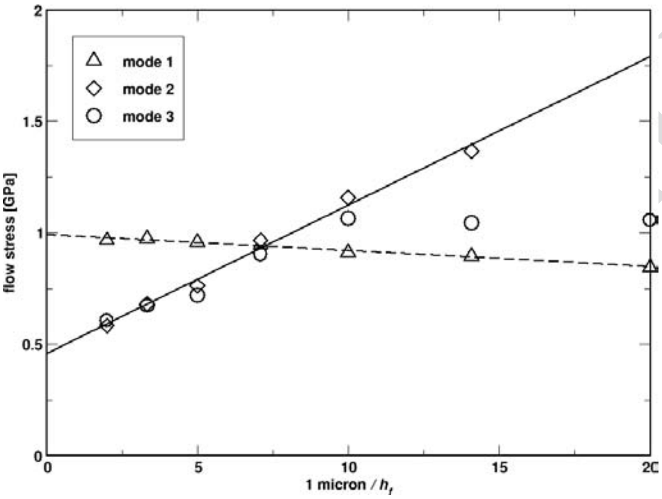

Fig. 4. Flow stress versus inverse film thickness for different deformation mechanisms as given in the legend.

4. Discussion

The present two-dimensional discrete dislocation dynamics study of plasticity in thin films shows that both diffusional creep and dislocation glide mechanisms are needed to account for the experimentally observed transition between the regime of a flow stress inversely proportional to the film thickness for micron to submicron thin films and the regime when the flow stress becomes independent of the film thickness for ultra-thin films below 400 nm thickness. The simulations reveal 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, as shown before.[12] The flow stress obtained from relaxation by dislocation slip on inclined slip planes shows a linear dependence on the inverse film thickness. The combination of both deformation mechanisms 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 can effectively shut down the creep mechanism, which results in slightly higher values of flow stress than those expected for pure creep deformation.

As seen in Figure 3, the linear scaling of the flow stress with inverse film thickness strongly depends on the initial stresses. The reason for the linear increase of the flow stress in geometrical confinement lies in the back stress of the dislocations piling up against the film substrate interface. Each active source will produce a number of dislocations until the local stress at the source position falls below the critical value. This occurs by virtue of the back stress induced by the pile up even if the average stress level in the film is still above the critical value for dislocation nucleation. If the initial stress is just above the critical stress for dislocation nucleation, only a single dislocation dipole can be generated by each source. The expansion of the dislocation dipoles produces plastic strain relaxation which reduces the average stress in the film below the critical value for dislocation nucleation. The final flow stress after relaxation is given by the number of dislocation dipoles that has been generated and by their maximum expansion in the film geometry. It is noted here that such strict linear scaling with inverse film thickness has not been observed in other work on dislocation modeling of thin film plasticity.[15]

The validity of the two-dimensional model for thin film plasticity is briefly discussed below. From fully three-dimensional models[8] as well as from *in-situ* transmission electron microscopy,[2,6,7] it is known that geometrical confinement of plasticity causes a strong curvature of the dislocations channeling through thin films or through obstacle forests. The motion of curved dislocations defines an intrinsic strength of the confined material systems. These effects are not captured within the two-dimensional model presented here. In this regard, 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 that are beneficial for plasticity, the geometrical confinement has a pronounced influence on the flow stress due to kinematic hardening caused by the back-stress of dislocations pile-ups. Such kinematic hardening causes the flow stress to depend strongly on the initial stress level from which the relaxation occurs. Experiments, in contrast, show that the room temperature flow stress is independent from the maximum temperature and thus insensitive to the initial stress level.[4, 5] This is probably due to thermal relaxation processes resolving the dislocation pile-ups in time.

This setback in the two-dimensional description of dislocation slip on inclined planes in the thin film geometry does, however, not affect the description of dislocation climb and dislocation slip on parallel slip planes. In this case, the dislocation nucleation criterion has been derived from fundamental simulations and the occurrence of multiple slip on parallel glide planes is in good agreement with experiment.[2] Furthermore, line tension effects can be considered to be small in this case. Thus, the main conclusion that the flow stress in thin films is limited by a transition from slip on inclined slip planes to diffusional creep is still considered valid, in particular since the linear scaling of the flow stress with the inverse film thickness in the conventional deformation regime is experimentally very well confirmed.

Finally, we note that the proposed hierarchical simulation approach appears to be a good strategy to model deformation in nanostructured materials. The key advantage of the mesoscopic modeling technique is that it allows reaching the length and time scales of laboratory experiments that are usually not accessible by atomistic simulations. The dependence of such systems on empirical parameters can be considered a drawback, however, with a careful design of the models these parameters can be given a physical meaning and their validation by fundamental simulation methods or by experiments becomes feasible. The possibility to purposefully include or exclude specific deformation mechanisms within the simulations in combination with comparison to experiment allows drawing substantial conclusions on the dominant deformation mechanisms in real structures.

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