STRENGTH AND DEFORMATION OF A THIN POLYCRYSTALLINE FILM ON A SUBSTRATE: DISLOCATION DENSITY ANALYSIS

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Abstract

The constitutive law of metallic polycrystalline thin films is determined on the basis of the consideration of the evolution, accumulation and formation of dislocations in thin film/substrate systems, taking into account the formation of misfit dislocations. An equation for the evolution of the dislocation density and the constitutive law for a thin polycrystalline metallic film are derived on the basis of the dislocation-density-related approach to the constitutive modeling of materials, taking into account the formation of misfit dislocations due to the threading dislocations movement, nucleation and multiplication of dislocations due to the interaction between misfit dislocations (like the Hagen-Strunk mechanism) as well as the effect of the misfit dislocations on the moving dislocations. It is shown that yield stress is proportional to a square root of a linear function of $1/L$, where L is the smallest characteristic length in a film (either thickness or grain size).

Introduction

Thin films present a key element in many devices, as integrated circuits, magnetic storage media, thermal sensing elements, optical filters, parts with protective (anticorrosion) coatings, etc. The performances and reliability of these and other thin film/substrate structures are determined to a large extent by the strength and fracture resistance of thin films on substrates.

The purpose of this work is to determine the constitutive law of metallic thin films on the basis of the consideration of the evolution, accumulation and formation of dislocations in thin film/substrate systems, taking into account the formation of misfit dislocations.

The deformation behavior of metallic thin films on substrates is influenced by the dimensional constraints on the dislocation movement [1], interface effects and microstructure of the film (grain boundaries, etc.) [2]. The effect of the film thickness on the strength of the film has been studied in many works. Table 1 shows several relations between the yield stress and the film thickness.

Table 1. Relations between the Yield Stress, and the Grain Size and Film Thickness

Relation between the yield stress and film thickness	Author	Approach
(1/h) ln h,	[3]	Analysis of the energy changes related with the extension of a misfit dislocation.
(1/h) (ln h + f)	[4]	Generalization of the model by Freund for the case of the motion of a mixed (edge/screw) dislocation, and for to the films on substrates under biaxial stress.
1/h	[1]	The stress necessary for yielding by dislocations channeling mechanism is determined from the condition that a dislocation loop fits inside the film
$\sqrt{K_{HP}^2/d + \tau_{source}^2}$ (while a smaller dimension, either grain size or film thickness, controls the flow stress),	[5-8]	Hall-Petch model [5], discrete dislocation simulation method [6-8] (in three dimensions).
A/h+B/d	[9]	Comparison of the the work done by a slip and the energy of going a dislocation loop along the sides and the bottom of the grain, in a polycristalline film.

Evolution of dislocation density and deformation of thin films

In order to derive a constitutive law for a thin film, we assume a power-law kinetic equation, relating the equivalent plastic strain rate and the equivalent stress [10, 11]:

$$\dot{\varepsilon}^p = \dot{\varepsilon}_0 (\sigma/\hat{\sigma})^m \tag{1}$$

where $\dot{\varepsilon}^p$ - equivalent plastic strain, $\dot{\varepsilon}_0$ and m – material parameters, σ and $\hat{\sigma}$ - equivalent

stress and internal variable of the material state. A stress to move a dislocation past two obstacles (in this case, the film/substrate interface and the free surface or oxide layer) is $\tau=\alpha\mu b/l=\alpha\mu b\rho^{1/2}$, where b – magnitude of the dislocation Burgers vector, l – distance between obstacles, μ=MG, M- average Taylor factor, G – shear modulus, α – a coefficient, $\rho\sim 1^{1/2}$ – dislocation density. According to Estrin [10, 11], the equation has a general validity. It is "obvious from dimensional considerations that the glide resistance must be given by Gb/L, where L – characteristic obstacle spacing in the glide plane". Von Blackenhagen [12] stated, however, that it is unclear, whether this equation is applicable for the dislocations in thin films, if the film thickness is of the same order as the Burger vector of the dislocations. Assuming that the film thickness is much more than the Burgers vector of a dislocation, we use this equation in our analysis.

Now consider the main factors influencing the dislocation density evolution in the metallic film:

- immobilization of dilocations at the film/substrate interface, grain boundaries and free surface; annihilation of dislocations stored at impenetrable obstacles,

- nucleation and multiplication of dislocations due to the interaction between misfit dislocations (like the Hagen-Strunk mechanism, when two crossing misfit dislocations annihilate locally and produce 2 new mobile dislocations, as well as other mechanisms),

- other misfit dislocation effects (like the formation of misfit dislocations due to the threading dislocations movement).

Following [10], the shear strain increment $M d\varepsilon^p$ can be determined in terms of "dislocation density increment $d\rho$ associated with immobilization of mobile dislocations at impenetrable obstacles", as the film/substrate interface and the free surface:

$$d\rho / d\varepsilon^p = (M/b)\sqrt{\rho} \qquad (2)$$

Romanov et al. [13] considered the evolution of the dislocation density in films in the framework of the analogy between the reaction kinetics in chemical systems and the dislocation evolution. They derived a similar equation for the source term in the differential equation for mobile dislocations in [13]: from $d\rho/dt = A\sigma(x) + ...$, and assuming $\sigma = \alpha\mu b\sqrt{\rho}$, one obtains $d\rho/dt = A(M/b)\sqrt{\rho} + ...$
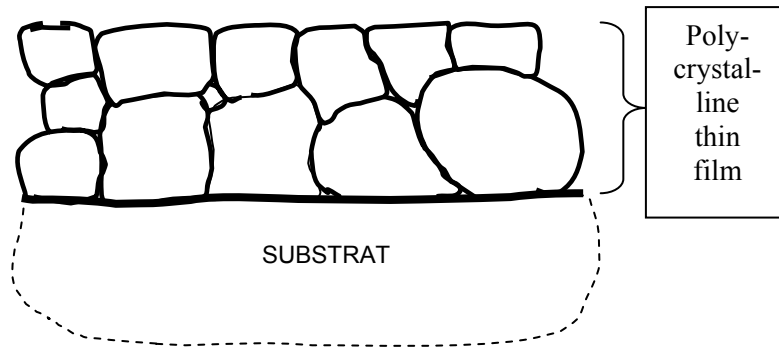

FIGURE 1. Thin polycrystalline film on a substrate. The grains are much smaller than the film thickness**.**

The mean free path L can be in this case identified with the film thickness, or grain size. In order to take into account the annihilation of dislocations stored at impenetrable obstacles (interface and free surface), we introduce the second term $-M k_2 \rho$, and have:

$$d\rho / d\varepsilon^p = M(1/bL - k_2 \rho), \qquad (3)$$

where L=d, k_2- recovery coefficient.

The multiplication of dislocations (by the Hagen-Strunk mechanism, or by other mechanisms) can be taken into account by adding the term with a breeding factor, using the model by Nix [4]:

$$d\rho/dt = \rho g v + \dot{\rho}_0, \tag{3}$$

where g – breeding factor (a value of the order of 5...20 dislocations/mm [4]), $\dot{\rho}_0$ - rate of nucleation of new dislocations in the substrate, v – dislocation velocity, $v = B \exp(C \tau_{eff}^{1/2})$, $C = (U/kT)\tau_0$, U – activation energy for the dislocation motion, $\tau_{eff} = \tau - W_{disl}/(bh/\sin\varphi)$, φ - an angle defining the slip plane normal, (φ can be taken 54°, see [4]), W_{disl} - work required to form a unit length of dislocation (a function of the film thickness, Burgers vector and shear moduli of the film and the substrate). If one neglects $\dot{\rho}_0$, one may obtain: $d\rho/d\varepsilon^p = (\rho g v/\dot{\varepsilon}_0)(\hat{\sigma}/\sigma)^m$.

The formation of misfit dislocations due to the threading dislocations movement can be described in the framework of the model by Nix [4]: $d\rho_{mf}/dt = v\rho$, where ρ_{mf} is the density of misfit dislocations.

- the stress governing the threading dislocation rate in thin films, is lower than the analogous stress in bulk material, due to the formation of the misfit dislocations,

- effect of the misfit dislocations on the moving dislocations.

The second effect is considered by substituting τ_{eff} for the stress τ in all the equations, where $\tau_{eff} = \tau - W_{disl}/(bh/\sin\varphi)$.

In [13], the "non-linear term" which accounts for the creation of climbing dislocations due to the gliding of threading dislocations, looks after some simplifications, as the term $-c\rho$ in an differential equation for $d\rho/dt$. (It is assumed that the climbing dislocations are transformed to the misfit dislocations when the reach the interface. The formation of the misfit dislocations by direct reaction between threading dislocation and interface is neglected.) Also, the blocking of threading dislocations by the misfit dislocations is neglected here.

The final equation for the dislocation density looks like:

$$d\rho/d\varepsilon^p = M(1/bL - k_2\rho) + (\rho g v + \dot{\rho}_0)/\dot{\varepsilon}^p = M[1/bL - \rho \, k_{20} \, (\dot{\varepsilon}^p/\dot{\varepsilon}_0^*)^{-1/n}] + (\rho g v + \dot{\rho}_0)/\dot{\varepsilon}^p, \tag{4}$$

or

$$d\rho/d\varepsilon^p = a_1 - a_2\rho, \text{ and } \sigma = a_3\sqrt{\rho} \tag{5}$$

where $a_1 = M/bL + \dot{\rho}_0/\dot{\varepsilon}^p$, $a_2 = M k_2 - gv/\dot{\varepsilon}^p$, and $a_3 = \alpha\mu b(\dot{\varepsilon}^p/\dot{\varepsilon}_0)^{1/m}$.

Assuming that the plastic strain rate $\dot{\varepsilon}^p$ =const and v=const, and integrating the equation, one obtains a formula for the dislocation density in a film:

$$\rho = (a_1 - \exp[a_2 (C-\varepsilon^p)])/a_2,\qquad\qquad(6)$$

and the constitutive law:

$$\sigma = a_3 [a_1 - \exp(a_2(C - \varepsilon^p))/a_2]^{1/2},\qquad\qquad(7)$$

The equation (7) can be rewritten in the form:

$$\sigma = (b_1/L - b_2)^{1/2},\qquad\qquad(8)$$

where $b_1 = a_3 M/b$, $b_2 = a_3 \exp(a_2(C - \varepsilon_p))/a_2]^{1/2}$. The derived constitutive law for the thin polycrystalline films is similar to the equation derived by Friedman and Chrzan [5], and confirmed numerically by von Blackenhagen et al. [6-8]

References:

1. E. Artzt, *Acta Mater*, 46, 16, (1988) 5611-5626

2. L. Mishnaevsky Jr and D. Gross, *Applied Mechanics Reviews,* 2003 (accepted)

3. L. B. Freund, *Adv. Appl. Mech.,* 30, (1994) 1-66

4. W.D. Nix (1989) *Met. Trans. A* 20A, 2217

5. L. H. Friedman and D. C. Chrzan. *Philosophical Magazine* (1998) *A* 77: 1185-1204.

6. B. von Blanckenhagen, P. Gumbsch and E. Arzt, Dislocation Sources in Discrete Dislocation Simulations of Thin Film Plasticity and the Hall-Petch Relation, Modelling and Simulation in Materials Science and Engineering 9 (2001) 157-169

7. G. Dehm, T.J. Balk, B. Von Blanckenhagen, P. Gumbsch, E. Arzt , Dislocation Dynamics in Sub-Micron Confinement: Recent Progress in Cu Thin Film Plasticity, Z. Metallkunde 93, No. 5 (2002) 383-391

8. Hartmaier, P. Gumbsch, E. Arzt, and W. D. Nix, Modeling Work Hardening in Thin Metal Films on Substrates, in ``*Werkstoffwoche* '98, Bd. VI'' edt. by R. Kopp et al. (Wiley-VCH Weinheim, Germany, 1999) 541-546

9. C.V. Thompson, *Journal of Materials Research* 8, 237, (1992)

10. Y. Estrin, In: *Unified Constitutive Laws of Plastic Deformation*, eds A. S. Krausz, K. Krausz, New York City, Academic Press, Inc., 1: pp 69-106 (1996)

11. Y. Estrin, In: *Multiscale Phenomena in Plasticity,* Ed.: J. Lepinoux et al, Kluwer Academic Publishers, (2000), pp. 339 – 348

12. B. von Blanckenhagen, Versetzungen in dünnen Metallschichten, MPI für Metallforschung, Stuttgart, 2002

13. A. E. Romanov et al., *Appl. Phys. Lett.* 69 (22), 1996