Stress Distributions and Thin Film Mechanical Properties

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Non-uniform stresses exist near the edges of thin films as a result of satisfying the boundary conditions. Growth stresses also contribute to stress gradients within the film, especially in structures with a void-columnar microstructure. As a result the strain-sensitive properties also must show spatial variations which will become more important as the specimen size decreases. We examine the origin of these distributions within the limits of elastic behavior, and discuss cases for which the distributions have also been measured. The role of plastic deformation in relaxation and fracture is introduced.

INTRODUCTION

Materials have been produced in thin film form for well over a century and their properties, which often apparently change from the corresponding bulk form, have been the subject of much intrigue. The availability of high and ultra high vacuum together with modern instrumentation for structure determination of films and surfaces has produced a flourish of literature in recent decades. Although much has been learned about specific film systems our knowledge is far from complete in matters of predicting spatial property changes and predicting failure modes and lifetimes. Furthermore, increasing surface to volume ratios must arise as we shift to submicrometre technology and will bring even tighter limitations to the production of thin film structures that must stand the rigors of the device world. It is the purpose of this paper to examine a number of considerations dealing with the mechanical properties of thin films. Furthermore, it is attempted to point out areas of emerging importance which possess as yet little fundamental information.

ORIGIN OF THE STRESSES IN DEPOSITED THIN FILMS

Large internal stresses develop in thin films as a result of the conditions of the condensation process and post-deposition treatments. These stresses result from satisfying the mechanical equilibrium of the film deposited on its substrate. Techniques for the measurement of the stresses and a tabulation of data are available in several reviews.[1] The purpose here is to recall that the fundamental reason these stresses develop is because there is a constrained volume change taking place in the film following its mechanical anchoring to the substrate. Usually a substrate is thick compared to the film giving rise to the stress distribution (Fig. 1). Unless otherwise stated in this paper it is assumed that the stresses and the elastic constants are homogeneous and isotropic; obvious but mathematically complicated expressions can be derived for the appropriate single crystal case

If the process giving rise to the stress is homogeneous then the condition known as plane stress arises (Fig. 1) in which the specimen has no forces normal to its plane and a uniform two-dimensional stress in the plane. Note that there is related isotropic strain in the plane plus a Poisson strain in the perpendicular direction. As usual in problems of elasticity the stresses and strain are connected by an elastic constant. In this case the appropriate equation is

$$\sigma = \frac{E}{1-\nu}\varepsilon$$

where σ represents the isotropic stress, ε the isotropic strain, E Young's modulus and ν Poisson's ratio for the film material.

Under the condition of elastic equilibrium, the stress distribution in both the film and substrate may be calculated. It is important to recognize that both a longitudinal strain and a bending will take place. With no external forces the neutral plane (plane of zero strain) is at a position two-thirds of the substrate thickness away from the interface when the film thickness is small compared to the substrate thickness.[1] When making such calculations one assumes no plastic deformation at the interface and the strain is continuous across the interface while the stress has a discontinuity. In practice, one must make certain that the introduction of plastic deformation or a fracture in either the film or substrate enters the stress analysis or interpretation of the data in a known way.

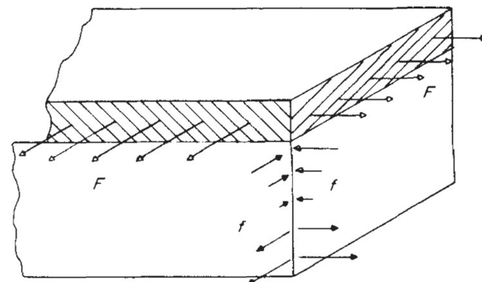

Figure 1. Stress distribution for a thin film in tension on rigid substrate. The integral of the forces vanishes for mechanical

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The physical origin of the magnitude of the stresses may be most easily understood by thinking of the corresponding strain which generates the stress. It may be a distributive process such as thermal expansion or a more localised process. Differential thermal expansion stresses may be calculated from the thermal expansion coefficients which are fortunately isotropic for cubic single crystals and polycrystalline materials. Differential thermal expansion strains are reversible so long as no relaxation takes place. Indeed, such a use of temperature induced stresses may allow the determination of onset of flow processes as well as the measurement of elastic constants or temperature coefficients for film materials.[2] Anomalous values of thermal expansion upon the initial heating of a system generally are evidence for recovery processes in the film rather than abnormal expansion coefficients. To oversimplify, the thermally induced stresses are commonly dominant in the cases of very high temperature depositions having a thermal mismatch, but can also be overwhelming in the case of very low melting point materials such as lead when cycled to low temperatures as is shown in a very detailed series of papers by Murakami[3] and others. In these papers the stress observed in the film is generally smaller than that calculated from the expansion coefficients. This difference arises because of the recovery processes in the specimens which in turn are related to the grain size and microstructure of the lead film. This work, perhaps the most detailed study of the relaxation processes in a film substrate system, emphasizes the importance of orientation effects, dislocation motion, and diffusion during the recovery.

The so-called intrinsic stresses, which should perhaps more properly be thought of as growth stresses, arise from different constraints which occur during film nucleation and growth. In Fig. 2 primarily those processes which are active during the deposition process itself are shown and should be considered as illustrative rather than all inclusive. As long as the atomic process involves a scale small compared to the film thickness, or perhaps the technique involved in the stress measurement, a homogeneous macrostress is observed as long as the mechanism is constant with film growth. Such a process is indicated in Fig. 3(a). Of course spatially localized regions, commonly the film/substrate interface or the outermost layers of the film, may harbor additional contributions. As regards the observed distributions, an interfacial contribution would appear [Fig. 3(b)]. The literature often presents the thickness dependence in three ways: the force per unit width, the average stress, and the stress distribution. For a stress generating mechanism localized at an interface, the average stress decays somewhat hyperbolically. Note that it is useful to carry out a dynamic measurement in which the stress is measured as a function of thickness in order to most

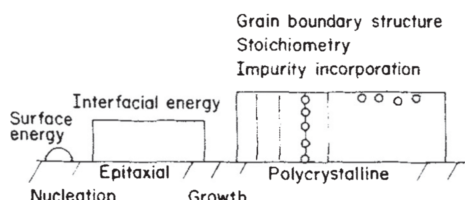

Figure 2. Physical origin of intrinsic stresses during nucleation

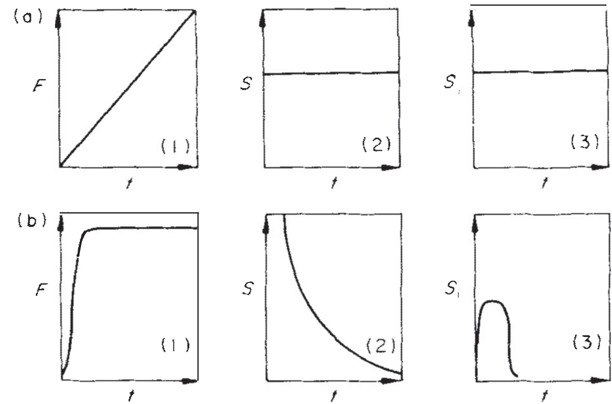

Figure 3. (a) Volume and (b) interfacial contributions to the three distributions commonly reported in the literature. (1) Force F; (2) average stress, $S = F/t$; and (3) instantaneous stress, $S_i = dF/dt$. t is film thickness.

easily identify the physical mechanism involved in the origin of the stress.

The goal to understand the physical origin of the large growth stresses is best reached through a knowledge of the film microstructure followed by semiquantitative concepts for the constrained volume change. Kinetics may be introduced crudely and are consistent with the observation that the growth stresses are small for temperatures at which the film atoms have considerable mobility ($>T_m/3$, T_m is melt temperature). As indicated in Fig. 2, surface and interfacial energies dominate during discontinuous[4] and epitaxial growth while grain boundary structures are most important for thick polycrystalline films.

The largest stresses are about 1% of the elastic modulus, and are found in medium to high melting point metals condensed on substrates held at temperatures $<T_m/4$. For 200 nm Ni films, the stresses are ~1 GPa tensile[5] in polycrystalline films with ~40 nm diameter columnar grains, but larger grain sizes and oxygen impurities lower the observed stress.[6] No stress relaxation is observed near room temperature. The force-thickness curve rises linearly giving confirmation to the volume generated stress mechanism of Fig. 3. A close examination of such data in the island growth stage shows a dependence on substrate material, presumably as a result of nucleation differences.

Growth stress tension is attributed to a film shrinkage during growth and commonly found for metallic deposits. Several microstructural models have been proposed for the origin of these stresses.[1] Ion implantation, gas entrapment during sputtering, or atomic peening during magnetron sputtering[7] are additional mechanisms which give rise to compressive stresses. It is suggested that the recent literature be consulted for the details for a particular technique or material. However the growth stresses often exceed those of thermal origin and lead to a driving force for mass transport and failure if they are not controlled.

Figure 4 indicates the contributions to stress changes arising from a post deposition treatment. The foremost one corresponds to a recovery or recrystallization when the annealing temperatures are sufficiently high ($>T_m/3$). The large grain boundary volume in polycrystalline films provides ready paths for mass flow.

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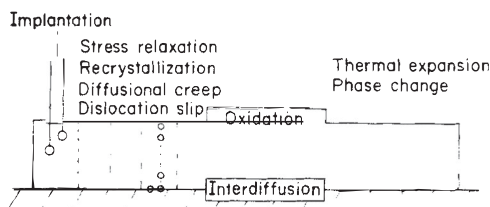

Figure 4. Typical mechanisms resulting in stress changes after film deposition.

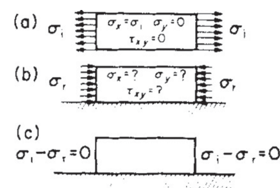

Figure 5. The boundary traction problem of a thin film under uniform intrinsic stress constrained to a rigid substrate.

recent attention as the origin of work softening of b.c.c. bulk materials.[8]

Thus the total stress distribution in a film/substrate package will be the sum of contributions from differential thermal expansion, growth stresses, and subsequent processing. The detailed contributions are commonly not known for a given system, although a given property will depend only on the total stress. Our knowledge of the relaxation processes is so poor that it is only known in a few instances[3] whether observed strains are determined by the stress production mechanism or essentially flow stress limited by the relaxation.

The objective of a simple quantitative model for the total stress in polycrystalline films is partially reached for the case of relatively pure metals when no relaxation is found. The role of impurities and initial growth stages have a qualitative explanation in terms of the microstructure. Dielectric films generally have a porous structure resulting in a sensitivity to the atmosphere with both reversible and irreversible stress changes following exposure to room ambient.

STRESS DISTRIBUTION

In this section the spatial variation of the stress in the plane of the film is considered, although of necessity the dimension perpendicular to the plane must also be examined. The variation of the stress near the edges of the film or within a particular grain of a continuous sample is calculated. The importance of stress gradients near the film edges and its relationship to the 'adhesion' has been considered previously.[9] However, since the cross-section of individual units in submicrometre structures are more apt to be square than planar, the importance of stress distributions in extremely small samples is presently of major importance.

For simplicity we consider that the mechanism giving rise to the stress is uniform. Because no forces are applied at the edge of the film, the free edge deforms so as to support the interior forces. We anticipate that there will be a characteristic distance for which the elastic distortion takes place, and the one-dimensional calculation shows that this distance is essentially the film thickness.

In order to understand the manner in which a stressed thin film transmits forces to its substrate, a continuum elasticity approach is taken. A thin film of thickness t is deposited on a substrate, and a very narrow cross-sectional slab is sliced from the coated substrate. The side view of the slab is one of a rectangular plate of length L and height t attached along its long edge to what shall be assumed as a semi-infinite, rigid medium.

strate thickness is usually at least 1000 times thicker than the film.

Bending plate experiments show that the condensed thin film possesses a large intrinsic stress, σ_i, that lies in the plane of the film, isotropic, is nearly uniform throughout the thickness, and is uniform over most of the area of the film. Consequently, a slab of thin film attached to its substrate is under a state of stress nearly identical to that of a rectangular plate that is clamped along an edge and that has undergone thermal expansion. The latter situation has been treated approximately by Aleck.[10] According to Aleck the problem is first converted to one of boundary tractions. The film is detached from the substrate and allowed to relax. To bring it back into a uniform state of intrinsic stress one imagines applying a uniform normal stress of magnitude σ_i to the ends of the film as shown in Fig. 5(a). Next, one imagines attaching the film back to the substrate and applying a uniform compressive stress σ_r to the ends, which in this case is equal in magnitude to σ_i because at the edge of the film there are no net applied normal boundary tractions. Consequently, one arrives back at the original situation which is depicted in Fig. 5(c) where $\sigma_i - \sigma_r = 0$. The problem is therefore one of finding the state of stress for the situation depicted in Fig. 5(b) and adding to it the state of stress depicted in Fig. 5(a). Note that the x direction is parallel to the film plane and the y direction normal to that plane.

Aleck solved the case for the stresses along the film/substrate interface for $L \gg t$. The curves plotted in Fig. 6 are based on his result. The origin is at the edge of the film. The pertinent features are (1) σ_x is just equal to σ_i until one gets within a distance t of the edge, (2) a large shear ($>2\sigma_i$) lies within a distance t of the edge, (3) a concentrated stress normal to the interface of magnitude

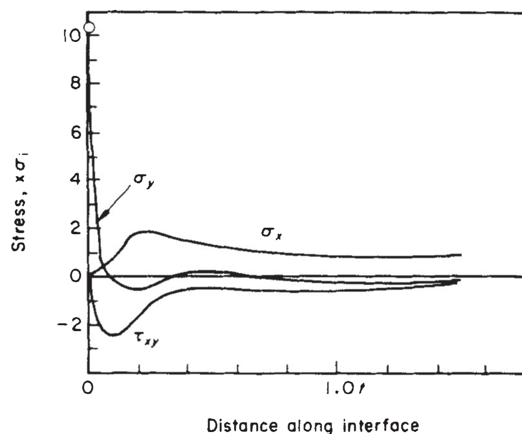

Figure 6. Normal and shear stresses near the free edge of a film. σ_x is in the plane and τ_{xy} the only shear component. The units are

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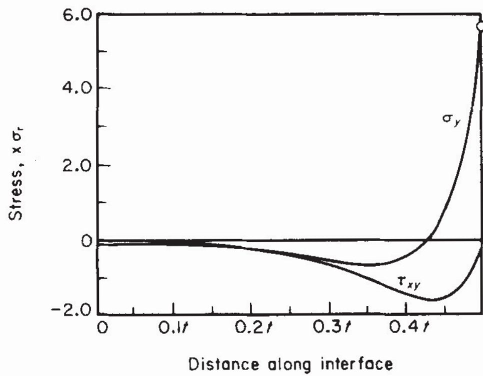

Figure 7. The interfacial stresses for a grain whose length equals its thickness and has a uniform stress applied to its boundaries normal to the substrate plane.

of about $10\sigma_i$ lies at the edge, and (4) there exist no non-zero shear stresses or stresses normal to the interface once at a distance somewhat larger than t from the edge. The value of σ_y normal to the plane at the film edge is an estimate since the elastic equations have a singularity at that point. Recent calculations by I. A. Blech (personal communication) indicate the solutions[10] are qualitatively correct, but too large by a factor of ~2–5. Practically, plastic deformation or fracture may take place to limit the stresses. For films that possess a tension intrinsic stress (σ_i greater than about 0.1 GPa) film 'adhesion failure' can occur at sharp edges of the film or defects in the film that are produced by scratching or fracture. This phenomenon is indeed one that is observed. Additionally, no such failure is expected to initiate at locations interior to the film that are more than a few film thicknesses away from any edge. We may also calculate the stress distribution within a single grain of a continuous polycrystalline film. A boundary stress σ_i, produced by the interatomic forces across a grain boundary during growth, is used to calculate the stress distribution within the isotropic grain. The boundary stress σ_r shown in Fig. 5(b) is equal to $-\sigma_i$ and no stress sources are present within the grain. The curves plotted in Fig. 7 are the interfacial stresses calculated from this geometry. The origin is placed at the center of the grain and $L = t$. Large forces are found near the edge of the grain if σ_i is appreciable. Since σ_r is $-\sigma_i$ for this case, the interfacial normal stress σ_y acts in a direction opposing film/substrate cleavage. The grain boundary forces lead

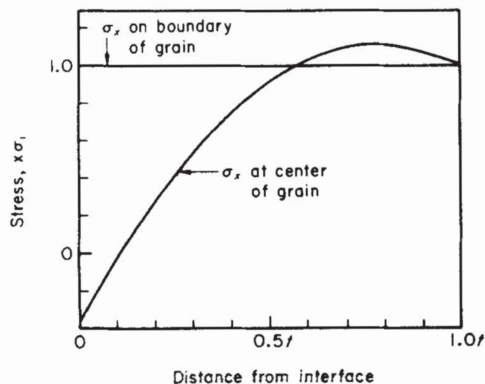

Figure 8. The distribution of stress parallel to the substrate plane through the film thickness in a grain whose length equals its

to similar shear stresses localized at the boundaries that can lead to plastic deformation at the film/substrate interface.

The variation of the elastic stress σ_x with the distance from the substrate is shown in Fig. 8 for the same grain at the grain boundary and at the center. The σ_x distribution changes continuously from $\sigma_x = \sigma_i$ at the boundary to the one shown in the figure at the center of the grain. The figure demonstrates that the stress in the plane of the film has the largest spatial variation near the substrate. In planes farther from the interface than half the film thickness, the planar stress is essentially constant.

Similar strain profiles have been determined on the basis of an exponential elastic relaxation near the edges[3] for isolated grains of different sizes. When the grain diameter is comparable to the thickness a sizeable portion of a given crystallite has large stress gradients. The thickness dependence of the stress relaxation in Pb films is related to the crystallite aspect ratio.

Strain gradients in the substrate may also be calculated. Blech and Mieran[11] have calculated that a region of lattice expansion is followed by lattice contraction in the surface region of a silicon wafer as one moves away from an edge of a window etched in the steam grown oxide layer under compression. The important point is that the strain gradients are localized (~50 film thicknesses) near the film edge. Lattice rotation, expansion, and contraction all occur, and X-ray scattering is enhanced. This distribution has recently been confirmed by 0.01 mm spatial resolution lattice curvature measurements by X-ray topographic techniques.

Chow[18] has treated the case of an external traction applied to an elastic substrate with a brittle film. Similar conclusions are reached; namely, the stresses are nearly constant except near the film edges. Since Chow essentially assumes the internal stresses are zero, there are several differences from the deposited film case, for which plane stress is found, σ_y is small except near the edges, and the neutral plane is not at the geometric center when longitudinal forces are considered in addition to pure bending.

INTERFACIAL FRACTURE

The preceeding elastic calculations confirm what experience has already taught us: the edge of the film or a crack resulting from a defect or film fracture forms an especially favorable spot for the beginning of failure. The propagation of a crack, either as a result of the large normal forces for a film under tension or the delamination that may take place because a film under

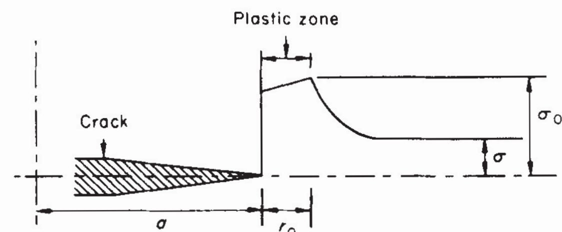

Figure 9. Stress distribution in front of growing crack for a

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compression is too large for the substrate, becomes the province of fracture mechanics.

For brittle materials, the Griffith concept has been successfully applied by Matthews and Klokholm[13] to analyze the cracking of magnetic bubble films. For a given applied stress, one failure mode took place when the film thickness exceeded the Griffith crack length. Chow[18] has successfully applied fracture concepts to the case of a brittle film on a stretched elastic substrate.

The fracture toughness approach incorporates the effect of plastic deformation near the tip of the propagating fracture as indicated in Fig. 9. Briefly, the approach defines a material constant which can be used to predict the critical stress which must be applied for a crack to propagate rapidly. The stress state and failure mode are important in the quantitative relations. The critical region is the stress peak near the growing crack tip, and the onset of plastic flow actually reduces the stress concentration at that point. If the highest stress that the material can sustain is the flow stress, a relation of the form $K = Y\sigma a^{1/2}$ may be used, where K is the material fracture toughness parameter, Y a proportionality factor which is geometrically dependent, σ the applied stress, and a the half-crack length. When the elastic energy released by the crack growth exceeds that absorbed by the plastic flow ahead of the crack, a rapid, and often catastrophic, failure results.

It seems obvious that this approach should be applied to thin film failure as it has in adhesion.[14,19] However, no thin film data in which the fracture toughness approach has been evaluated to give a useful parameterization are known to the author. Often it is not known if the crack propagation is taking place in the film, or substrate, or at the interface on an atomic scale. Furthermore, the mechanical properties of the material comprising the interfacial region are not those of either bulk film or substrate. Cases of brittle intermetallic compounds and large stress gradients are known.[15] It is suggested that progress will come when the fracture toughness approach is extended to the film geometry. Direct tensile measurements on micrometre size specimens are now possible following the development of the nanotensilometer[16] and applications to film materials will follow. The challenge to produce higher strength materials for specialized purposes by structure modulation is within sight.[17]

SUMMARY

Qualitatively, we already have an approach to understanding failure in film structures maintained at modest temperatures and environments. The total film stress must be determined first. If it is less than ~0.1 MPa generally no problems are anticipated. If it is in the gigapascal range, especially if brittle intermetallic compounds are formed at the film/substrate interface, failure is rapid when a critical thickness is reached and the forces can no longer be supported across the interface. Fracture is possible in either the film or substrate material. High temperatures with dislocation motion or the diffusion of impurities along the interface, perhaps driven by the stresses, may lead to a slow failure. Our predictive capacity is not good here; for a longer time scale where fatigue and creep are important we have no literature background to date.

We can summarize the present knowledge of mechanical properties of thin films as follows:

1. Elastic moduli and thermal expansion coefficients are the same as a bulk phase of the same composition and microstructure with the exception of the surface region.
2. Large intrinsic stresses are developed during film growth. They are subject to some control by deposition technique and process variables through the microstructure. Thermal stresses often dominate at high temperatures.
3. The stresses and stress gradients serve as a driving force for relaxation and property inhomogeneities in addition to the better known catastrophic failure.

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