NANOINDENTATION HARDNESS OF SOFT FILMS ON HARD SUBSTRATES: EFFECTS OF THE SUBSTRATE

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ABSTRACT

The ability to accurately measure the mechanical properties of thin metallic films is important in the semiconductor industry as it relates to device reliability issues. One popular technique for measuring thin film mechanical properties is nanoindentation. This technique has the advantage of being able to measure properties such as hardness and elastic modulus without removing a film from its substrate. However, according to a widely-held rule of thumb, intrinsic film properties can be measured in a manner which is not influenced by the substrate only if the indentation depth is kept to less than 10% of the film thickness, which is often not practical. In this work, a method for making substrate independent hardness measurements of soft metallic films on hard substrates is proposed. The primary issue to be addressed is the substrate-induced enhancement of indentation pile-up and the ways in which this pile-up influences the contact area determined from analyses of nanoindentation load-displacement data. Based on experimental observations of soft aluminum films on silicon, glass, and sapphire substrates, a simple empirical relationship is derived which relates the amount of pile-up to the contact depth. From this relationship, a simple method is developed which allows the intrinsic hardness of the film to be measured by nanoindentation methods even when the indenter penetrates through the film into the substrate.

INTRODUCTION

It has recently been demonstrated that indentation pile-up has important influences on the nanoindentation measurement of the hardness and elastic modulus of monolithic materials [1]. In order to obtain accurate results, the extra contact area generated by the pile-up around the hardness impression must be incorporated into the analysis procedures. Recent research has also shown that the amount of indentation pile-up in soft films on hard substrates can be greater than that in monolithic materials due to constraints on plasticity in the film imposed by the substrate [2, 3]. The amount of pile-up in these films varies with indentation depth, reaching a peak value when the maximum indentation depth, h_{max}, is approximately twice the film thickness, t_f. Due to the inability of current nanoindentation analysis procedures to account for pile-up in the determination of contact areas, the nanoindentation hardness, H_{nano}, and elastic modulus, E_{nano}, can significantly overestimate the true values [2,5]. In one recent study, the nanoindentation hardness of a very soft aluminum film on a hard glass substrate was found to overestimate the actual hardness by as much as 100% [5]. It was also found that when the extra contact area due to pile-up was taken into account, the hardness of the film for indenter penetration depths less than the film thickness was essentially independent of depth; only when the indenter penetrated through the film into the hard substrate did the actual hardness begin to increase.

In this study, aluminum films deposited on several hard substrates materials were tested by nanoindentation methods to examine how the amount of pile-up in a soft film deposited on a hard substrate is influenced by the nature of substrate material. Based on the experimental observations, a simple method is developed which can be used to correct nanoindentation hardness measurements for the influences of pile-up and thereby determine

the intrinsic hardness of the film even when the indenter penetrates through the film into the substrate.

EXPERIMENTAL PROCEDURE

Several thin films of aluminum were deposited on three different hard substrate materials using two different deposition techniques. Physical vapor deposition was used to obtain aluminum films on silicon. The Al/Si system was chosen due to its widespread application in the semiconductor industry and because many models for substrate influences on hardness have been developed based on data obtained for this system [3,6-8]. Films of four different thicknesses, t_f = 240, 500, 640, and 1700 nm were studied. Magnetron sputtering was used to deposit aluminum films on soda-lime glass and (100) sapphire. These sputtered films were prepared simultaneously to a thickness of 500 nm. Relevant substrate properties as measured by nanoindentation techniques are listed in Table 1, where the subscripts "s" and "f" refer to the substrate and film, respectively.

Table 1. Thin film specimens tested in this research.

Film Material	Substrate Material	H_s/H_f	E_s (GPa)	H_s (GPa)
Aluminum	Silicon	12	160	12
Aluminum	Glass	6.5	70	6.5
Aluminum	(100) Sapphire	26	450	26

The specimens were tested in a nanoindentation system using a well-calibrated Berkovich diamond indenter. The load and displacement data obtained in the nanoindentation tests were analyzed according to the method of Oliver and Pharr [4] to determine both the nanoindentation contact area (A_{nano}) and the nanoindentation hardness (H_{nano}). Results obtained from these analyses were compared with actual indentation contact areas (A_{actual}) measured from scanning electron micrographs (SEM), and actual hardnesses (H_{actual}) determined by dividing the peak indentation load, P_{max}, by A_{actual}. Care was taken in the measurement of the actual contact area to include the area contained in the pile-up, since this area can contribute significantly to the load-bearing capacity of the contact. The amount of indentation pile-up was characterized by the ratio A_{actual}/A_{cc}, where A_{cc} is the corner-to-corner area of the indentation, that is, the area of the triangle defined by the positions of the indentation corners as observed in the SEM images. As discussed elsewhere [2], A_{actual}/A_{cc} provides a useful measure of the amount of pile-up when there is little or no pile-up at indentation corners, as was the case for most of the indentations in this work. The value A_{actual}/A_{cc} = 1 implies that there is no pile-up (or sink-in), while values greater than 1 correspond to proportionally larger amounts of pile-up.

RESULTS AND DISCUSSION

Figure 1 shows for the dependence of the nanoindentation hardness, H_{nano}, on the indenter penetration depth normalized with respect to the film thickness, h_{max}/t_f. Included in the plot are data for each of the three different film/substrate systems. At small depths, the hardnesses of all three specimens converge to values between 0.8 GPa and 0.9 GPa, while at large depths, the hardnesses increase rapidly toward the values of their respective substrates. Note that the hardnesses of all three materials are very similar at all depths less than the film thickness. This result was somewhat surprising since the hardnesses of the substrates vary over a such a wide range, i.e., from a low of 6.5 GPa for the glass to a high of 26 GPa for the sapphire (see Table 1).

Using the actual contact areas measured in SEM images for hardness computations, the actual hardnesses, H_{actual}, for each of the 500 nm films are shown in Figure 2. The most significant difference between the data in Figures 1 and 2 is the relative magnitude of the hardnesses for indentation depths beyond the film thickness; close inspection of the two plots

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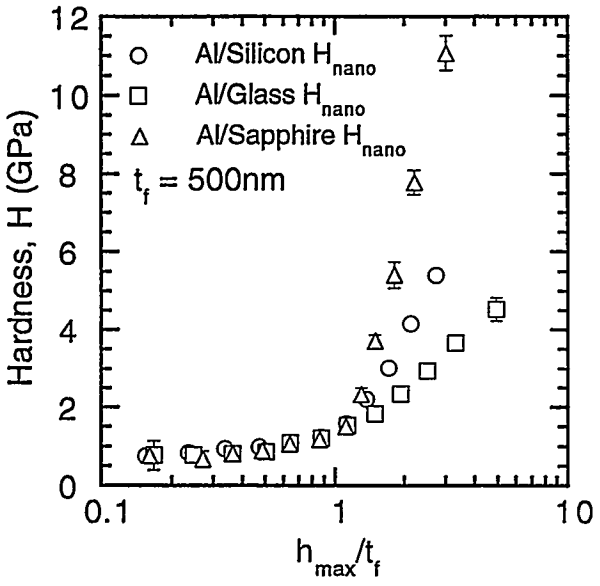

Figure 1. Nanoindentation hardness of 500 nm aluminum films.

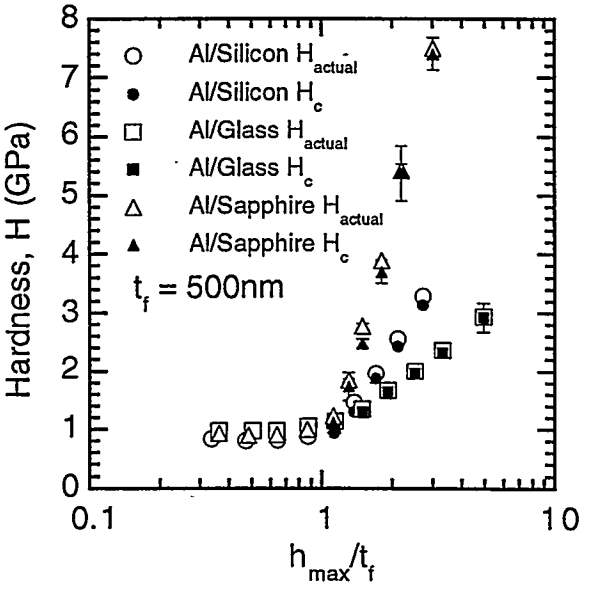

Figure 2. Indentation depth dependence of the hardness of the 500 nm films

shows that the actual hardnesses are much less than those predicted by the nanoindentation procedures. The reason for this difference is that the nanoindentation analysis ignores pile-up, which is enhanced in a thin-film/hard-substrate system when the indenter penetrates to a significant fraction of the film thickness. Predictions of a model for the composite hardness, H_c, of a film/substrate system developed by Tsui, Oliver, and Pharr [2] are also plotted in Figure 2. The parameters used to compute the composite hardness were H_f = 1.0 GPa, H_s = 6.5 GPa (glass), and H_s = 26 GPa (sapphire), α = 0.78 (glass), and α = 0.80 (sapphire). The procedures used to apply the composite hardness model will be discussed later in this paper. Note that the predictions of the model are in good agreement with the experimental data.

In order to examine the structure of the pile-up, an indentation made in the 1700 nm Al/glass specimen was cross-sectioned and imaged by using a focused ion beam technique. Figures 3a and 3b show cross-sections perpendicular to and parallel to one of the indentation edges. It is evident that the film remains fully-adherent after indentation, i.e., there is no

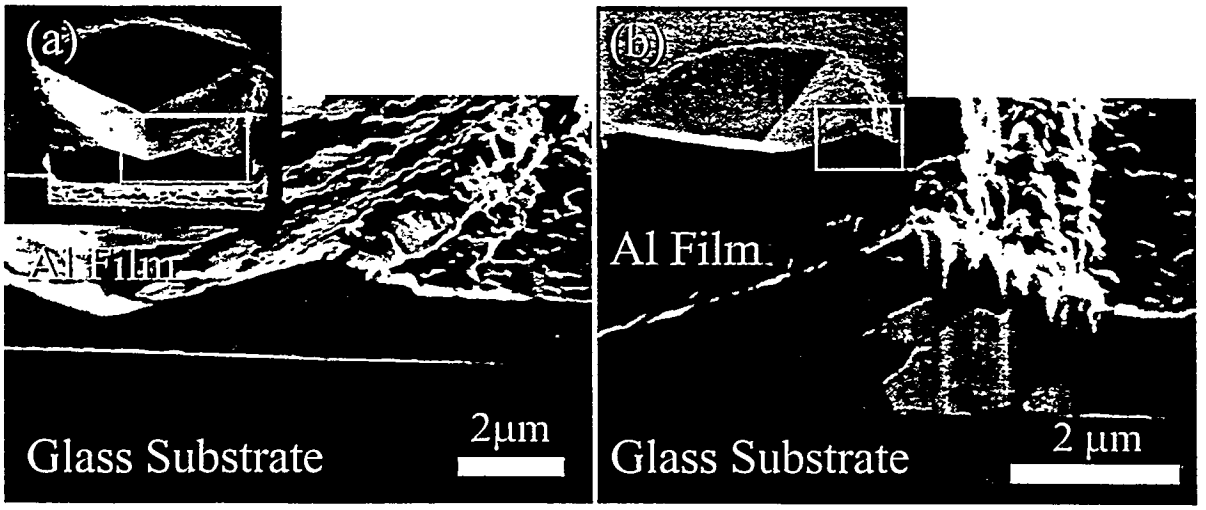

Figure 3. Focused ion beam images of indentation cross-sections : (a) prependicular to and (b) parallel to one of the indentation edges.

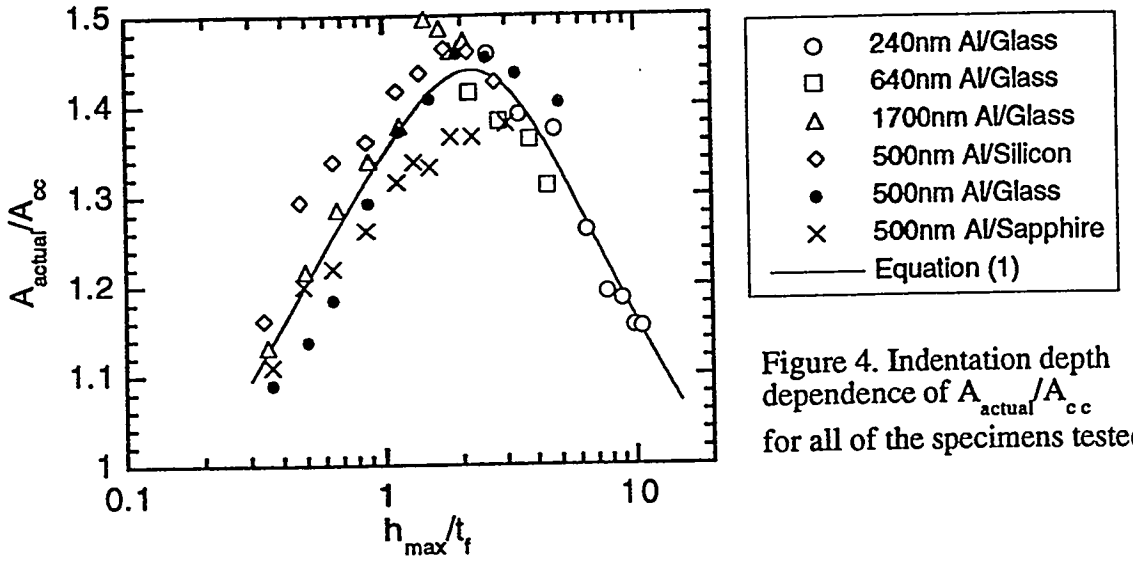

Figure 4. Indentation depth dependence of A_{actual}/A_{cc} for all of the specimens tested.

evidence of delamination in the vicinity of the hardness impression. The grain structure in the film is also visible, and a curious observation is that while the grains beneath the indentation are relatively equiaxed, those outside it are more columnar.

Figure 4 illustrates an important characteristic of the pile-up behavior. Plotted in this figure is the ratio A_{actual}/A_{cc}, i.e., the ratio of the actual contact area measured from SEM images to the corner-to-corner area, as a function of the penetration depth, h_{max}/t_f. For each film, the ratio is close to 1.1 at small depths, indicating only a small amount of pile-up, but rises with increasing depth until it passes through a maximum at $h_{max}/t_f \approx 2$. The maximum value is in the range 1.4-1.5, indicating that 40-50% of the contact area at these depths is due to pile-up. At larger depths, the ratio decreases because a greater portion of the hardness impression resides in the hard substrate. The substrate materials are not prone to pile-up during indentation but rather sink-in.

An important feature of the data in Figure 4 is how the A_{actual}/A_{cc} values group tightly around a single master curve, even though the data include results for films produced by two very different deposition methods and three different substrates with greatly different hardnesses and elastic moduli (see Table 1). This suggests that the film pile-up behavior, at least for the substrate materials examined in this study and possibly others, can be described by a simple, substrate independent relationship between A_{actual}/A_{cc} and h_{max}/t_f. Clearly, however, such a function cannot apply to all substrates. For example, if the substrate were made of a material with mechanical properties similar to those of aluminum, the system would behave not as a film on a substrate but rather as a monolithic material for which A_{actual}/A_{cc} must be depth independent due to the geometric similarity of the Berkovich indenter. It seems reasonable, however, that a universal relationship may hold for substrates which are significantly harder than the film, since above some critical relative hardness ratio, H_s/H_f, one would expect no appreciable plasticity in the substrate until the indenter penetrates through the film. For these materials, the substrate would behave like a rigid foundation, with the pile-up behavior determined primarily by the plastic deformation characteristics of the film material. Since the softest substrate used in this study was glass with H = 6.5 GPa, the critical hardness ratio must be less than $H_s/H_f = 6.5$. Curve fitting procedures showed that in the range $0.3<h_{max}/t_f<10$ the relationship between A_{actual}/A_{cc} and h_{max}/t_f is reasonably described by the empirical relation :

$$\frac{A_{actual}}{A_{cc}} = 1.905\left(\frac{h_{max}}{t_f}\right)^{-0.210} - 1.260\exp\left(-1.095\left(\frac{h_{max}}{t_f}\right)\right) - 0.135\left(\frac{h_{max}}{t_f}\right)^{-1} \qquad (1)$$

Equation 1 has been plotted as the solid line through the data in Figure 4.

Based on the observation of substrate independent pile-up behavior, it is possible to develop a new nanoindentation procedure for measuring the hardness, H_f, of a soft film on a hard substrate which accounts for the influences of the substrate and pile-up. The method may thus be used to provide a measurement of the intrinsic hardness of the film. Because it is empirically based, the procedure, as developed here, formally applies only to aluminum films deposited on those substrates for which $H_s/H_f > 6.5$, but it may also prove useful for other soft film materials such as gold and copper and for softer substrate materials.

The procedure is based on the composite hardness model for a soft-film/hard-substrate system developed by Tsui, Oliver , and Pharr [2]. As described elsewhere [2], the model suggests that the composite hardness, H_c, for an indentation made either entirely in the film or which penetrates into the substrate, can be estimated from a simple area fraction approximation:

$$H_c = \left(\frac{A_f}{A_{actual}}\right)H_f + \left(\frac{A_s}{A_{actual}}\right)H_s \qquad (2)$$

where A_f and A_s are the portions of the projected indentation contact areas in the film and substrate, respectively, and $A_{actual} = A_f + A_s$ is the total indentation contact area. Noting that $H_c = H_{actual}$, this equation can be re-written as:

$$H_f = \left[H_{actual} - \left(\frac{A_s}{A_{actual}}\right)H_s\right]\left(\frac{A_{actual}}{A_f}\right) \qquad (3)$$

from which it follows that the intrinsic hardness of the film can be determined from measurements of the parameters on the right hand side of the equation. Using procedures described below, each of these parameters can be conveniently measured from nanoindentation load-displacement data alone, that is, without having to image the indentation.

The measurements require two separate indentations: one in the bare substrate and the other in the film/substrate system. From the indentation in the substrate, standard nanoindentation analysis procedures can be used to determine the hardness of the substrate, H_s, and a parameter $\alpha = h_c/h_{max}$, where h_c is the contact depth and h_{max} is the maximum depth of penetration. With these parameters, A_f and A_s can then be evaluated from the film/substrate nanoindentation data by assuming the interface between the film and substrate sinks-in to produce the same deflection geometry that would occur if there were no film on the substrate. As discussed elsewhere [2], this assumption is reasonable as long as $H_f << H_s$ and/or $h_{max} > t_f$. With this assumption, the depth, h_s, along which contact is made between the indenter and the substrate can be estimated from, $h_s = \alpha\,(h_{max} - t_f)$. Once h_s is established, A_s follows by evaluating the area function of the indenter at h_s, and A_f can be computed from $A_f = A_{actual} - A_s$.

To complete the evaluation of the intrinsic film hardness, the value of A_{actual} on the right hand side of Equation 3 is computed from Equation 1 by assuming that $A_{cc}=A_{nano}$, that is, the corner-to-corner area is reasonably well-estimated by the area deduced from the nanoindentation load-displacement data of the film/substrate indentation. This assumption works well provided there is no pile-up at the corners of the indentation, which was verified in this study by SEM investigation of the hardness impressions. After computing H_{actual} from P_{max} and A_{actual}, all of the quantities needed for the evaluation of H_f from Equation 3 are known.

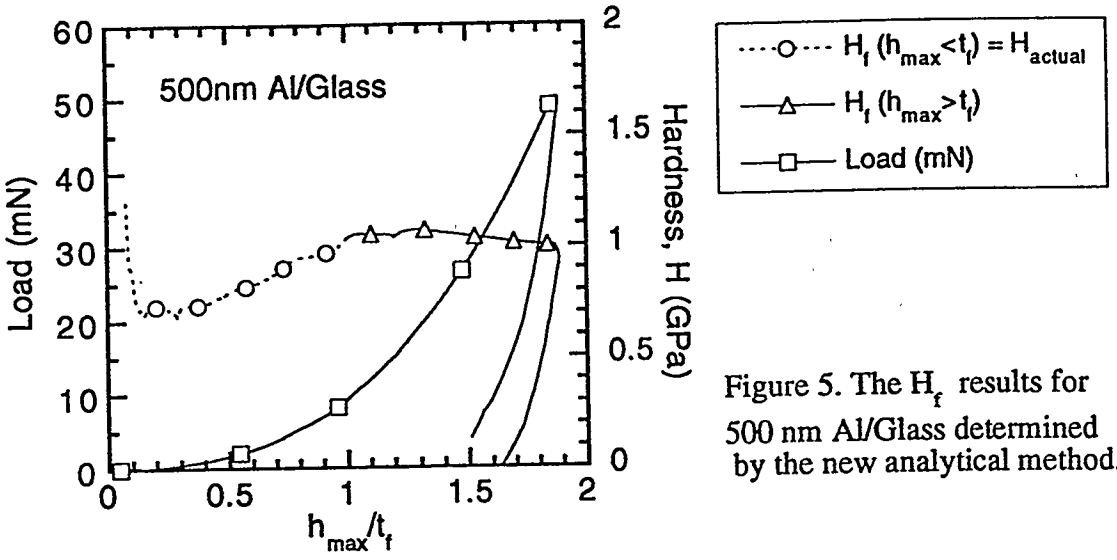

Figure 5. The H_f results for 500 nm Al/Glass determined by the new analytical method.

To illustrate its utility, the new procedure was applied by making an indentation in the Al/glass specimen using the continuous stiffness measurement technique (CSM) to obtain the nanoindentation data [4]. With continuous stiffness measurement, all the quantities needed to compute the film hardness can be measured continuously as a function of depth as a single indentation is made. Separate measurements of the bare glass substrate yielded $\alpha = 0.78$ and $H_S = 6.5$ GPa, which were used in calculations.

Figure 5 shows the film hardness evaluated by the new procedure as a function of h_{max}/t_f. Note that the hardness is relatively independent of depth with a value just under 1 GPa, in good agreement with what is expected based on the small depth data in Figure 2. Thus, with the new procedures, a reasonable estimate of the intrinsic film hardness may be obtained at any of a variety of indentation depths, including indentations which penetrate through the film into the substrate. The technique could prove valuable for measuring the intrinsic hardness of films which cannot be conveniently indented without penetrating the substrate, e.g., very thin films.

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