PHASE ANGLE IN INDENTATION-INDUCED DELAMINATION WITH BUCKLING AND ITS APPLICATION TO INDENTATION DELAMINATION IN ZnO FILM/SI SUBSTRATE SYSTEMS

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ABSTRACT

A formula is derived to calculate the phase angle of interfacial mixed mode fracture in the indentation delamination test, in which the delaminated film buckles. Interfacial fracture toughness depends on the phase angle and calculating the phase angle is necessary for the determination of mode I interfacial fracture toughness.

ZnO films with different thicknesses were deposited on 525 μm-thick (100) Si substrates. Then, Vickers indentation tests were carried out on the ZnO/Si systems at room temperature. For the systems with film thicknesses being equal to and thicker than 0.832 μm, only indentation-induced delamination occurred when indentation loads were low. Under high indentation loads, radial cracking concurrently occurred with delamination. The radial cracks were invisible at the film surfaces because the crack length was smaller than the delamination size. Combining the composite hardness models with the indentation-induced delamination model, we developed a method to determine the interfacial fracture energy between a film and its substrate. The novel method is particularly useful for indentation equipment without any displacement measurement devices. Using the new method, we extracted the interfacial fracture energy to be about 12.2 J m^{-2} and from 9.2 to 11.7 J m^{-2} for the cases without and with buckling of delaminated films, respectively. Consequently, the pure mode I interfacial fracture energy was calculated to be 10.4 J m^{-2} for the ZnO/Si systems.

1 INTRODUCTION

Thin films on substrates have been attracting intensive interests from academic and industrial researchers due to their wide applications in the fields such as microelectronics, optoelectronics and microelectromechanics etc. The durability and reliability of devices composed of thin films on substrates depend greatly upon the mechanical properties of film/substrate systems. One of the most important mechanical properties is adhesion between a film and its substrate. When the adhesion is evaluated by fracture, it is more accurate to call it the interfacial fracture toughness. Due to the small dimension of film thickness, it is a challenging task to accurately measure the interfacial fracture toughness between a film and its substrate. Researchers are developing new techniques to characterize the interfacial fracture toughness in film/substrate systems and six developed testing methods are briefly summarized in a recent overview [1]. Among the six testing methods, the indentation-induced delamination method is the most attractive approach because of its easiness and simplicity. Furthermore, indentation tests may be more appropriate than other methods when sample size and/or testing region become very small. In the indentation-induced delamination test, stresses near an interface crack tip are always comprised of both mode I and mode II components, thereby leading to a mixed mode fracture. The mode mixity is characterized by a phase angle, ψ. Measured interfacial fracture toughness depends on the phase angle. Jensen and Thouless [2] proposed the following empirical formula,

$$\Gamma(\psi) = \Gamma_I^c[1 + (1-\lambda)\tan^2\psi], \tag{1}$$

where $\Gamma(\psi)$ is the interfacial fracture toughness at a phase angle, ψ, $\Gamma_I^c(\psi = 0)$ is the pure mode I interfacial fracture toughness, and λ is a fitting parameter. Since $\Gamma(\psi)$ changes with the phase angle, Γ_I^c is the fundamental property to represent the adhesion strength between a film and its substrate. To accurately determine the value of Γ_I^c from measured $\Gamma(\psi)$, it is necessary to precisely determine the phase angle. Huang et al. [3] investigated the phase angle for the indentation delamination test and applied the theoretical results in the assessment of Γ_I^c between ZnO and Si [4]. The present work briefly reports the results published in References [3, 4]. Interested readers may refer References [3, 4] for detailed information.

2 PHASE ANGLE

The theoretical analysis is based on the thin film approximation that the film thickness is much smaller than the substrate thickness and considers only circular buckling. Following Hutchinson and Suo's approach [5], Huang et al. [3] derived the phase angle, which is given by

$$\tan\psi = \frac{\left[\dfrac{\sigma_I}{\sigma_B} - \dfrac{3.6304(1+\nu_f)}{\mu^2}\xi^2\right]\sin\varpi - \sqrt{12}\,\dfrac{4.2156}{\mu^2}\xi\cos\varpi}{\left[\dfrac{\sigma_I}{\sigma_B} - \dfrac{3.6304(1+\nu_f)}{\mu^2}\xi^2\right]\cos\varpi + \sqrt{12}\,\dfrac{4.2156}{\mu^2}\xi\sin\varpi}, \tag{2}$$

where σ_I denotes the indentation-induced compressive stress for axisymmetric indentation delamination, σ_B is the critical buckling stress, $\mu = 3.8317$, ξ is the ratio of the buckling amplitude at the center of the circular buckled film to the film thickness, and ϖ is a dimensionless function of Dundurs' elastic mismatch parameters. Under the thin film approximation, Suo and Hutchinson [6] calculated and tabulated the value of ϖ and $\varpi = 52.1°$ if the film has the same elastic constants as the substrate.

3 ENEGRY RELEASE RATE FOR INDENTATION-INDUCED DELAMINATION

Marshall and Evans [7] and Evans and Hutchinson [8] developed the fundamentals for measuring interfacial fracture energy from the indentation-induced delamination test. They modeled the delamination region as a clamped circular plate with a delamination radius, *a*, on a rigid substrate. During indentation testing on a film/substrate system, an indenter penetrates and displaces a volume, V_I, into the film, which induces a compressive stress, σ_I, in the film. The indentation-induced compressive stress is the driving force to delaminate the film from the substrate and the corresponding strain energy release rate, G, for the delamination is:

$$G = \frac{h_f\sigma_I^2(1-\nu_f^2)}{2E_f} + (1-\alpha)\frac{h_f\sigma_R^2(1-\nu_f)}{E_f} - (1-\alpha)\frac{h_f(\sigma_I-\sigma_B)^2(1-\nu_f)}{E_f}, \tag{3}$$

where E_f, ν_f, h_f, and σ_R are Young's modulus, Poisson's ratio, thickness and residual stress of the film, respectively. The parameter α in eqn (3) is given by

$$\alpha = 1, \text{ for } \sigma_I + \sigma_R < \sigma_B \text{ (no buckling)}, \tag{4a}$$

and

$$\alpha = 1 - \frac{1}{1 + 0.9021(1 - \nu_f)}, \text{ for } \sigma_I + \sigma_R > \sigma_B \text{ (with buckling)}. \tag{4b}$$

4 EXPERIMENTS

Four-inch (100) silicon wafers with a thickness of 525 μm were used as substrates, which were cleaned by HF-dipping before film deposition. The surface roughness of the substrates was measured to be less than one nanometer. A ZnO thin film was deposited on a silicon substrate in a magnetron sputtering system (Automated Research Coater, Plasma Science Inc., U.S.A.) with a 2-inch-diameter ZnO target of 99.99% purity. The pressure in the chamber was pumped down to 1×10^{-2} Torr prior to deposition with a ratio of Ar to O_2 to be 90 to 10. The substrate was mounted on a rotated plate, 30 cm away from the target. The sputtering deposition was conducted at room temperature with a power of 100 W. The thickness of each ZnO film was controlled by deposition time. A Tencor P-10 surface profiler was employed to measure the film thickness after deposition, wherein a small area of the film was removed by the standard photolithography technique to expose the original Si surface. The measured thicknesses of ZnO thin films are 0.202 ± 0.012, 0.283 ± 0.010, 0.443 ± 0.014, 0.554 ± 0.015, 0.832 ± 0.013, 1.103 ± 0.015, 1.192 ± 0.014, and 1.535 ± 0.015 μm, respectively. The roughness of the film surfaces, measured by the surface profiler, varies from 8 ± 1 nm to 21 ± 1 nm with a mean value of 14.5 ± 3.2 nm. The crystal quality of the ZnO films was studied by x-ray diffraction (XRD) with a Philips PW1830-based diffractometer. The radiation was generated from a Cu anode with the $K_{\alpha 1}$ wavelength of 1.540562 Å. The XRD pattern exhibits an intense peak for the (002) diffraction at 34.20°, comparable to the 34.335° expected for a bulk ZnO sample. The other peaks at 72.05° and 69.13° represent the diffractions from the ZnO (004) and the substrate Si (004), respectively. This result indicates that the film has all of its crystallites in the preferred orientation with the *c*-axis normal to the substrate surface.

A load-controlled microhardness tester (MHT-4, Anton Paar) was used to carry out indentation tests with a 136° pyramidal Vickers diamond indenter. The loading ability of the tester ranges from 0.5 mN to 2.0 N, whereas the employed loading range, in the present study, is from 10 mN to 2.0 N. The indentation test was conducted at a loading rate of 200 mN s^{-1} to reach a preset peak load, then held at the peak load for 10 seconds, followed by an unloading course. For each preset peak load, at least 10 tests were done at different sites, and a mean value of the tests was taken in following calculations and analysis. In all tests, the crystallographic orientation of the wafers was fixed, with the diagonals of the impression parallel to the <110> directions of the silicon wafers. The optical microscope of the microhardness tester was used to measure the delamination radius and the radial crack length. After the indentation tests, the delamination profile was measured with the Tencor P-10 surface profiler.

5 RESULTS AND DISCUSSION

In the ZnO/Si systems with the 1.103, 1.192 and 1.535 μm-thick films, delamination was the dominant damage mode over the entire load range. Figure 1 shows the evolution of the delamination with the indentation load for the 1.535 μm-thick-ZnO/Si system, wherein three characteristics describe the damage behavior. Firstly, the delamination radius, *a*, increased with the load if the load was lower than 0.75 N. When the load exceeded 0.75 N, the delamination radius remained unchanged as the indentation load increased. Secondly, buckling of the film occurred at the load of 0.45 N, which resulted in interference rings. Thirdly, there were several through-film-thickness cracks within the delamination region. The other systems with 1.103 and 1.192 μm-thick films also showed similar delamination behavior.

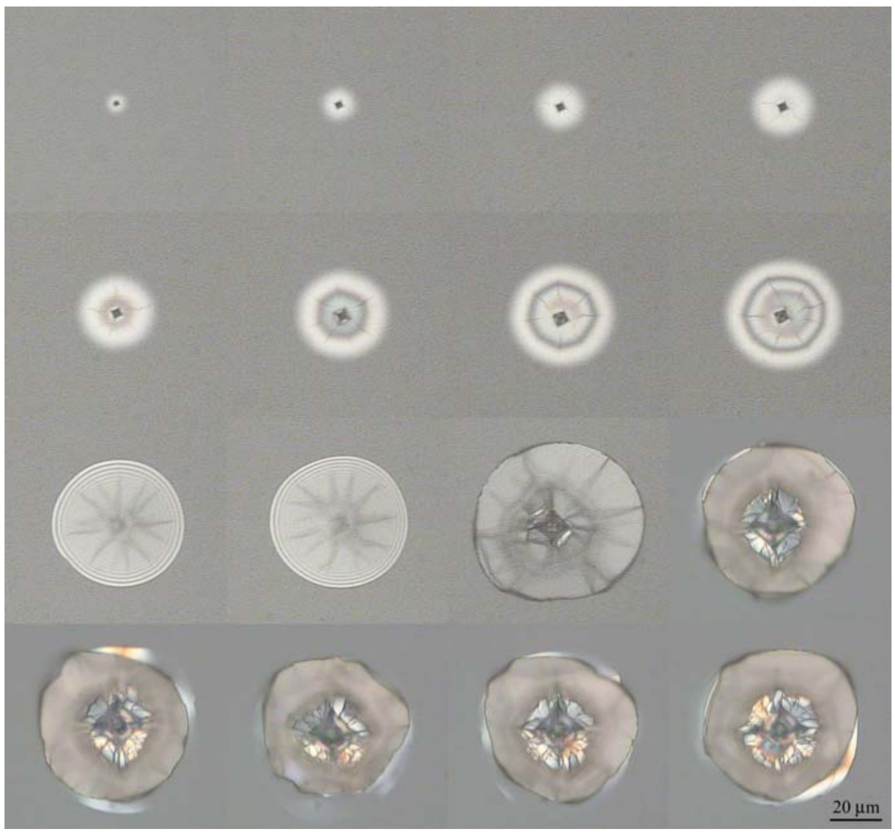

Figure 1: Optical photos to demonstrate the development of indentation-induced delamination in the 1.535 μm-thick ZnO/Si system after indented under 0.05, 0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.45, 0.50, 0.75, 1.00, 1.25, 1.50, 1.75 and 2.00 N, respectively.

As indicated in figure 1, there exists a critical load for buckling of the film. The critical loads for buckling of the 0.832, 1.103, 1.192 and 1.535 μm-thick films were found to be 250±10, 275±10, 295±10 and 420±10 mN, respectively. No buckling occurred when the applied load was smaller than the critical load for a given film thickness. The microscopic observations of the

1.535 μm-thick film indicated that for the tests that the load exceeds 430 mN, buckling occurred immediately after the indenter withdrew from the film. At the critical load of 420 mN, buckling occurred with a slight delay (1-5 minutes) after the indenter was completely unloaded. Figure 2 shows the development of the buckling for the 1.535 μm-thick film at the load of 420 mN. Comparing the delamination size, a = 23.43 μm, in figure 2(a) to the delamination size, a = 24.86 μm, in figure 2(b) illustrates that buckling increased the delamination size, thereby indicating the presence of compressive film stress. Buckling also caused the delaminated film to warp upward and finally to crack around the impression to form an internal circle. The topography of the buckled film thus looks like a volcano, where the internal circle may be compared to the rim of the crater.

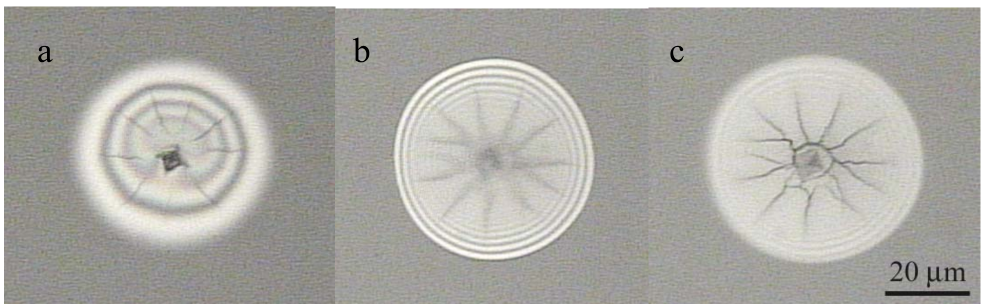

Figure 2: Optical photos to show the development of buckling of the 1.535 μm-thick-ZnO/Si system after indented under the critical load of 420 mN. (a) The state at about two minutes before buckling. (b) Final state after buckling. Focus on the perimeter of the debonded film. (c) Final state after buckling. Focus on the internal circle.

Experimentally we found that radial cracking occurred when the load was higher than 750 mN. The radial crack length in the substrate increases as the load increases from 750 mN to 2.0 N, whereas the delamination radius is almost independent of the load and remains unchanged over this load range. In addition, for a given indentation load, the thicker the film, the shorter the radial crack in the substrate is. This is attributed to the less indenter penetration into the substrates covered with the thick films and the depression effect induced by the compressive film stresses.

With eqn (3) and the measured delamination size, we calculated the critical energy release rate. The experimental data at the indentation depths smaller than the corresponding film thicknesses were used to extract the interfacial fracture energies, Γ_c. For the ZnO/Si system, we obtain ϖ= 51.7°, which is independent of the film thickness under the thin film approximation. Then, we calculated the phase angle, which varies from 2.6° to 13.5°, changing with the film thickness and the indentation load, when bucking occurs. Clearly, the phase angles with and without buckling show that there is a significant effect of mode II loading on the measured interfacial fracture energy. From the obtained interfacial fracture energies and phase angles and using eqn (1), we have Γ_I^c = 10.4 J m^{-2} and λ = 0.89 for the present ZnO/Si systems. The fitting curve is illustrated in Figure 3.

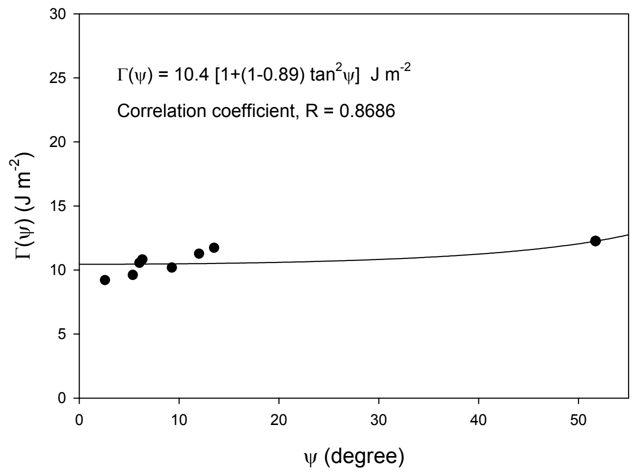

Figure 3: Fitting experimental data of the interfacial fracture toughness with the empirical formula, $\Gamma(\psi) = \Gamma_I^c[1+(1-\lambda)\tan^2\psi]$, to extract the value of Γ_I^c.

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