A hybrid experimental/numerical approach to characterize interfacial adhesion in multilayer low-κ thin film specimens

Rajesh Kitey a,c,1, Nancy R. Sottos b,c,* , Philippe H. Geubelle a,c

a Department of Aerospace Engineering, University of Illinois at Urbana-Champaign, IL 61801, USA
b Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, IL 61801, USA
c Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana-Champaign, IL 61801, USA

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ABSTRACT

The strength of multilayer low dielectric (low-κ) constant organo–silicate glass (OSG) and silicon–carbon–nitride (SiCN) thin film interfaces is characterized by a laser spallation technique. Two specimen sets with different OSG/SiCN stacking sequences are evaluated. The effect of helium (He) pretreatment is also investigated. The stress at the low-κ interface is enhanced through the use of a fused silica backing layer to shape the incident pulse and the addition of a thin gold (Au) top layer to increase inertial force during the dynamic failure event. The weakest interface in the multilayer stack (first to fail) is identified through optical images, profiometry and scanning electron microscope images of the spallation zone. The strength of the failed interface is inferred from the incident stress history in combination with a one dimensional dynamic wave propagation analysis. The adhesion strength of the OSG/SiCN interface (372 MPa) is 32% larger than that of Si/OSG (282 MPa) for specimens with no He pretreatment. The interfacial strength of both interfaces is significantly increased by a He pretreatment, making failure by spallation difficult.

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1. Introduction

New low dielectric (low-κ) materials are rapidly replacing conventional insulating materials in electronic packaging due to reduced size scale and superior performance [1]. The advantages of these material systems are also accompanied by mechanical reliability concerns. Delamination at low-κ film interfaces in multilayer stacked configurations is common when the semiconductor devices are subjected to thermo-mechanical loading. Hence, reliability of the device is dependent on the strength and integrity of the low-κ interface.

Characterization of the adhesion strength of low-κ thin film interfaces presents a significant experimental challenge. The layers are very thin, typically less than 300 nm, and sandwiched between multiple thin films in the device. Conventional test methods such as the four-point bend [2–4], indentation [5,6] and stud-pull [7] have limited application for low-κ multilayer devices. Hartfield et al. [1] and Dauskardt et al. [2] found that failure at some low-κ interfaces could not be induced in a four point bend experiment prior to cleavage of the Si substrate. Zheng et al. [8] characterized the work of adhesion of low-κ films by a scratch test. However, the measured adhesion energy was higher than that obtained from four-point bend test due to the influence of test parameters and sample geometry. Volinsky et al. [9,10] applied the nanoindentation technique to assess the interfacial properties but in most cases observed fracture in the low-κ films prior to interfacial delamination. Kong et al. [11] characterized the interfacial properties of black diamond and porous methyl-silsequioxane films with a wedge indentation technique. The film delamination behavior and the formation of corner cracks were dependent on the indenter angle. Using modified edge lift-off technique, Kim et al. [12] investigated the effects of chemical and physical structures on the adhesion performance of low-κ films. The interfacial fracture toughness was dependent upon the film thickness and the thickness of the epoxy backing layer. This dimension dependent fracture behavior was attributed to the change in morphology and the physical properties of the film.

In contrast to the quasi-static loading in most adhesion tests, the laser spallation technique [13–23] employs high amplitude laser-induced acoustic stress pulses to dynamically load a thin film interface, as shown schematically in Fig. 1. These pulses have sharp temporal rise and fall times and subject the thin film to strain rates of the order 107/s. The spallation technique was first introduced by Yang [13] and Vassen [14] and further developed by Gupta and co-workers [15] who introduced interferometric measurements for characterization of the thin film interfacial strength. The method was applied to characterize metallic, ceramic and polymer film interfaces [14–23] with the film thickness ranging from few hundred nanometers to a few microns. More recently, Kandula et al. [23] developed a laser
spallation protocol to quantify the critical adhesive strength of a tough poly-p-phenylenebenzobisoxazole polymer and silicon nitride (SixNy) interface in a multilayered thin film stack. Here, we further extend the laser spallation tool to characterize low-κ thin film interfaces with film thicknesses less than 300 nm.

A significant challenge to overcome when applying the laser spallation technique to very thin, strongly adhered interfaces is premature cleavage failure of the Si substrate, prior to interfacial failure. The laser pulse generates a high amplitude compressive stress wave in the substrate. This stress wave propagates through the specimen and upon reaching the free surface of the film, reflects back as a tensile wave. The resulting interface stress between the film and the substrate is dependent on the stress difference between the incident and reflected waves and the thickness of the film,

\[ \sigma_{\text{int}}(t) = \frac{2 \rho_{\text{film}} C_{\text{film}}}{\rho_{\text{sub}} C_{\text{sub}}} \left[ \sigma_{\text{sub}} \left( t + \frac{h_{\text{film}}}{C_{\text{film}}} \right) - \sigma_{\text{sub}} \left( t + \frac{h_{\text{film}}}{C_{\text{film}}} - h_{\text{film}} \right) \right] \approx \rho_{\text{film}} h_{\text{film}} \frac{\partial^2 u}{\partial t^2} \]

(1)

where \( \rho \) is the density, \( C \) is the dilatational wave speed and \( h \) is the film thickness. The subscripts sub and film represent substrate and thin film, respectively. For a given interface strength, adhesive failure of thinner films requires a stress pulse with either a larger magnitude or a sharper fall time. Simply increasing the magnitude of the incoming stress pulse (i.e. increasing the laser fluence), however, can result in cleavage failure of the Si substrate due to the reflected tensile wave prior to reaching the critical interface strength, as shown in Fig. 2.

In the current work, we introduce two methods to generate higher interface stresses without increasing the magnitude of the incoming pulse. Based on the work of Wang et al. [19–21], we instead alter the shape of the incoming pulse. The entire multilayer thin film specimen is bonded to a non-linear fused silica (FS) substrate, which generates a decompression shock with a more rapid fall time. In addition, we deposit a thin layer of gold (Au) on the top film layer of the specimen. The relatively high density of the Au effectively increases the dynamic inertial force acting on the thin film stack. Both modifications significantly enhance the interface stress generated during the spallation experiment, and enable characterization of several low-κ film specimens.

2. Materials and methods

2.1. Specimen preparation

The multilayer thin film samples consisted of two different stacking sequences of silicon-carbon-nitride (SiCN) and organo-silicate-glass...
(OSG) thin films on a <100> Si substrate are defined as Type 1 and Type 2 specimens in Table 1. For both configurations one set of specimens was pretreated in a helium (He) environment during processing. For specimens designated Type 1-A and Type 2-A in Table 1, an additional 250 nm thick Ti/Au film was deposited by E-Beam evaporation onto the top of the free low-κ film surface. The back surface of the Si substrate was bonded to a FS disk by applying a thin layer of UV-cured adhesive at the FS/Si interface. A 400 nm layer of aluminum (Al) was then deposited on the FS back surface by E-beam evaporation, followed by spin coating of a waterglass layer of ~6 μm thickness on top of the Al layer. The waterglass was spun at 2500 rpm for 3 min.

Additional calibration experiments were carried out on samples consisting only of a 30 nm Si3N4 passivation layer on the top of the Si substrate (Table 1). The back surface of the Si substrate was bonded to a FS plate as described above, followed by deposition of a 400 nm Al film and a 6 μm layer of waterglass on the back surface of the FS.

2.2. Spallation set-up

The laser spallation experiment is shown schematically in Fig. 1. An 8 ns duration Gaussian pulse of variable energy content (0–150 mJ) from the Q-switched Nd:YAG laser (λ = 1064 nm) is focused down to a circular spot of 1 mm diameter onto the constrained Al energy absorbing layer at the specimen back surface. If no FS is present, a compressive acoustic pulse develops in the Si substrate with shape similar to the Gaussian profile of the incident laser pulse. If the Si substrate is bonded to a FS plate as shown in Fig. 1b, the negative nonlinear elasticity of the FS causes the initial Gaussian pulse to evolve into a decompression shock with a 1–2 ns fall time. Reported previously by Wang et al. [19–21], this non-linear pulse shaping significantly enhances the interface stress for a substrate pulse. The compressive shock wave is transmitted across the FS/Si interface, reflecting back as a tensile wave upon reaching the free surface of the top inertial Ti/Au layer, subjecting the film interfaces to mode-I loading conditions. The laser energy is gradually increased to amplify the developed interfacial stresses until the weakest interface fails.

2.3. Interferometric measurements

Out-of-plane displacements are measured on the sample surface using a Michelson interferometer, shown schematically in Fig. 1 [20]. The variation of fringe intensity, I(t), at the point-of-interest is recorded using a 2 GHz LeCroy oscilloscope and related to the fringe order, N(t),

\[ I(t) = \frac{I_{\text{max}} + I_{\text{min}}}{2} + \frac{I_{\text{max}} - I_{\text{min}}}{2} \sin 2\pi N(t) \]  

where \( I_{\text{max}} \) and \( I_{\text{min}} \) are maximum and minimum intensities of the fringe in consideration, respectively. The out-of-plane displacement history, \( u(t) \), is calculated from the Doppler shift[24],

\[ u(t) = \frac{\lambda}{2} N(t). \]  

\[ \sigma_{\text{sub}}(t) = -\frac{1}{2} (\rho C_v)_{\text{sub}} \frac{\partial u}{\partial t}, \]

where \( \lambda \) is the wave length of the probe beam (514 nm). If the rise time of the substrate pulse is significantly smaller than the time required for the wave to propagate through the film, the following thin film assumption is valid for evaluating the substrate stress history [19],

\[ \sigma_{\text{sub}}(t) = -\frac{1}{2} (\rho C_v)_{\text{sub}} \frac{\partial u}{\partial t}. \]

Fig. 3. Interferometric measurements obtained from a Si/Si3N4 calibration experiment. (a) Displacement fringes recorded using photo-diode; (b) corresponding out-of-plane displacement history \( u(t) \); (c) the associated substrate stress history \( \sigma_{\text{sub}} \).
where $\rho$ and $C_d$ represent density and the dilatational wave speed, respectively.

Representative interferometric data from a calibration specimen are shown in Fig. 3. A sharp turn-around point at $-262$ ns in the fringe data (Fig. 3(a)) indicated the arrival of a decompression shock, which was transmitted across the FS/Si interface. The out-of-plane displacement history (Fig. 3(b)) at the $\text{Si}_N_x$ free surface was calculated by analyzing interferometric fringes in Fig. 3(a) using Eqs. (2) and (3). The displacement monotonically increases until the turning point is reached. The substrate stress, $\sigma_{\text{sub}}$, was evaluated from the displacement history by applying Eq. (4). The substrate stress history in Fig. 3(c) reveals the characteristic linear ramp of the decompression shock attaining a peak compressive value of 0.92 GPa followed by a sharp fall. Multiple wave reflections in a multilayer film stack complicate analytical calculations of the interface stress, leading us to adopt hybrid experimental/numerical approach to evaluate stress histories at multilayer film interfaces.

2.4. Dynamic wave propagation analysis

A one-dimensional finite element analysis was developed to study the wave propagation in the multilayer thin film specimens and analyze the stresses developed at each interface. The non-linear FS substrate was not explicitly included in the analysis. Instead, we imposed the substrate stress history measured directly from calibration experiments at the same fluence. The top and bottom surfaces of the multilayer film stack were assumed in plane strain with traction free boundary conditions. The specimen geometry was meshed using elements of size $h/20$ where $h$ is the thinnest layer dimension ($h = 30$ nm in current simulations). This element size was chosen based on a convergence study. An explicit direct integration scheme with a forward difference time steps was applied for transient analysis. The simulations were performed using a time step $\Delta t/5$ where $\Delta t$ is the minimum of wave travel time in the elements of considered layers.

2.5. Interface stress calculation

The interface stress evolution in the multilayer specimens was determined through a combined experimental and numerical approach shown schematically in Fig. 4. A series of laser spallation tests were conducted on FS/$\text{Si}/\text{Si}_N_x$ calibration specimens (Table 1) over a range of laser fluences. The substrate stress profile was calculated from interferometric measurements at each fluence (Fig. 3(c)). A series of laser spallation experiments were then conducted on Type 1-A and Type 2-A multilayer low-$\kappa$ specimens over a range of fluences. The critical laser fluence to initiate failure of weakest interface in the stacked configuration was identified from a combination of profilometric data, collected using a surface profiler (Dektak) with 2.5 $\mu$m stylus radius, and the images of the spallation zone were obtained with a scanning electron microscope (SEM, Hitachi S4700). The interface stress at this critical fluence was calculated numerically using the substrate stress profile measured

Table 2

<table>
<thead>
<tr>
<th>Layer</th>
<th>Density (kg/m$^3$)</th>
<th>Elastic modulus (GPa)</th>
<th>Poisson’s ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>FS</td>
<td>2200</td>
<td>73.5</td>
<td>0.17</td>
</tr>
<tr>
<td>Si$^a$</td>
<td>2330</td>
<td>125.0</td>
<td>0.29</td>
</tr>
<tr>
<td>OSG</td>
<td>1500</td>
<td>7.8</td>
<td>0.15</td>
</tr>
<tr>
<td>SiCN</td>
<td>1900</td>
<td>67.0</td>
<td>0.19</td>
</tr>
<tr>
<td>Ti</td>
<td>4510</td>
<td>102.7</td>
<td>0.33</td>
</tr>
<tr>
<td>Au</td>
<td>19,282</td>
<td>116.0</td>
<td>0.36</td>
</tr>
</tbody>
</table>

$^a$ Material properties in -1,0,0 direction.

Fig. 4. Schematic showing hybrid experimental/numerical approach to evaluate interfacial strength of multilayer low-$\kappa$ films.

Fig. 5. Effect of substrate stress profile and Ti/Au inertial layer on the developed interface stress between Si and OSG in a Type 1 specimen. (a) Transient loading histories used in the one dimensional elastodynamic simulation (stress profiles are evolved symmetric Gaussian stress pulse upon traveling $t_w$ ns in FS); (b) the stress history at the Si/OSG interface.
on the corresponding calibration sample. The material properties of various layers used in the experiments and computations are listed in Table 2 [25–27]. The low-κ layer (OSG and SiCN) properties are provided by Texas Instruments Inc.

3. Results and discussion

3.1. Effect of incident stress profile and inertial layer

The effect of the incident stress profile on the interfacial stresses in a Type 1 specimen was investigated numerically for two different loading profiles of same amplitude shown in Fig. 5(a). The substrate stress profiles were generated by using the stress-time relationship [21],

\[ \sigma(t) = \sigma_{\text{max}} \exp \left( -\frac{2t^2}{T^2} \right) \]

where \( \sigma_{\text{max}} \) is the maximum amplitude of compressive stress pulse, \( T \) is the fall time of the laser pulse and \( t_0 \) is given by

\[ t_0 = t - t_w \left( 1 - \frac{3\sigma}{\alpha t} \right) \]

The \( \sigma \) is stress at time \( t \), \( t_w \) is the wave travel time, and \( \alpha \) and \( \beta \) are the non-linear material constants of the pulse shaping material. The stress profiles in Fig. 5(a) were generated assuming FS material properties [28], \( \alpha = 77.6 \text{ GPa} \) and \( \beta = -415.9 \text{ GPa} \) and \( t_w = 0 \text{ ns} \) (profile 1) or \( t_w = 45 \text{ ns} \) (profile 2). The symmetric Gaussian pulse (profile 1) is representative of a stress wave developed in a Si substrate with no FS layer. Profile 2 is similar to the decompression shock wave achieved with the addition of a FS backing layer [21]. The resulting stress developed at the Si/OSG interface is plotted in Fig. 5(b) for both stress profiles. Only a small maximum tensile interface stress (13 MPa) is developed for profile 1. The maximum interface stress increases to 146 MPa for profile 2, indicating that the interface stress is significantly magnified by using a backing layer of pulse shaping material like FS.

The effect of a higher density inertial layer was also investigated for the Type 1 specimen configuration. The inertial layer consisted of a 25 nm thick Ti film and a 250 nm thick Au film deposited on top of the SiCN layer. Simulations were performed for both stress profiles and the resultant Si/OSG interface stresses plotted in Fig. 5(b). The inertial layer increased the interface stress to 179 MPa (dashed curve) compared to 13 MPa in the absence of Ti/Au layer (dotted curve) for profile 1. The increase in interface stress was more dramatic for profile 2 with a peak of 685 MPa, more than 4 times the interface stress developed in the absence of the inertial layer.

3.2. Interfacial failure

Failure at the weakest interface in a multilayer low-κ film specimen was initiated by loading the interfaces beyond critical laser fluence, necessary to initiate spallation. Interface failure was identified by optical and scanning electron microscopy in combination with surface profilometry in the damage zone. In all of the spallation experiments on as-received Type 1 and Type 2 specimens, cleavage of the Si occurred prior to interfacial failure (Fig. 2). As predicted by the numerical simulation described in Section 3.1, it was necessary to optimize the interface stress through the addition of a non-linear pulse-shaping substrate material (FS) and a Au top layer to increase inertia.

Representative images of interfacial failure in untreated and He pretreated Type 1-A specimens are shown in Fig. 6. The film lift-off at the onset of spallation initiation in Fig. 6(a) and (c) corresponds to the laser fluences 55 and 74 mJ/mm², respectively. As expected, the
interfacial damage increases with the increasing incident laser fluence in both untreated and He pretreated specimens. For the same laser fluence, the He pretreated specimen has a smaller damage zone compared to its untreated counterpart (Fig. 6(b) and (d)). Moreover, the failed interface (spalled region) appears more textured in He pretreated specimen in comparison to the untreated case. The dotted regions of failure around primary damage zone, indicative of the failure initiation, are more prominent in the untreated specimen (Fig. 6(b)). The untreated Type 1-A surface profile across the damaged region in Fig. 7(a) (solid curve) reveals a ca. 560 nm [29] deep spallation zone, indicating interfacial failure at the Si/OSG interface. In case of the He pretreated specimen the failure depth (dashed curve in Fig. 7(a)) varies between 500 and 550 nm, suggesting not only the Si/OSG interfacial failure but the cohesive fracture within OSG as well. The surface profile of the He pretreated specimen also shows greater roughness (20.0± 2.5 nm) than its untreated specimen counterpart (9.5±2.0 nm). The presence of rough surface in the damaged zone further supports that there is cohesive failure at the Si/OSG interface. Failure initiation at higher laser fluence and the resulting higher surface roughness at the failed interface indicates that He pretreatment increases the bond strength between the Si and OSG film interface.

The micrographs in Fig. 9 show representative interfacial damage for untreated and He pretreated Type 2-A specimens. Similar to Type 1-A, interfacial failure initiates at higher laser fluence in the He pretreated specimen (Fig. 9(c), Γ = 79 mJ/mm²) compared to its untreated (Fig. 9(a), Γ = 63 mJ/mm²) counterpart. The circumferential band within the damage zone of untreated Type 2-A specimen in Fig. 9(b) may be due to failure of multiple interfaces. The surface profile for this specimen in Fig. 7(b) (solid curve) reveals a ~530 nm deep damage zone that corresponds to the failed interface between OSG and SiCN, as well as an intermediate region at 275 nm that indicates debonding of the Ti/Au inertial layer deposited on the top of the OSG. Further evidence of multilayer interfacial failure is observed by SEM (Fig. 8(c)). The relatively smooth surface of the OSG top reveals interfacial debonding between the OSG and Ti/Au layers whereas the rough surface at the OSG/SiCN interface indicates interlayer cohesive failure. The absence of the SiCN layer also suggests that the interface between Si and SiCN could not be failed in current experimental investigation. In contrast, the He pretreated specimen displays a uniform smooth surface within the spallation zone (Fig. 9(d)). This trend is also evident from the spallation surface profile in Fig. 7(b) (dashed curve) and indicates that only the Ti/Au inertial layer is debonded during the spallation experiments. No interfacial failure was observed in any of the low-k film interfaces for the type 2-A He pretreated specimens.

3.3. Interface strength

A combined experimental and numerical scheme is applied to determine interlayer stresses at the incident laser fluence as described in Section 2.5. The effect of laser fluence on the substrate stress amplitude during the calibration experiment is shown in Fig. 10(a). For laser fluences ranging from 39 to 95 mJ/mm², the substrate stress monotonically increases from 0.26 to 1.17 GPa. The interfacial stresses are determined for each test specimen configuration using explicit finite element analysis and the interferometrically measured substrate stress history as the loading condition. The maximum stress calculated at the Si/OSG interface in Type 1-A specimens and the SiCN/OSG in Type 2-A specimens is plotted as a function of laser fluence in Fig. 10(b). The nearly overlapping interface stress curves monotonically increase from 100 to 550 MPa for the incident laser fluence varying between 39 and 95 mJ/mm². The interfacial stress corresponding to the laser fluence just before failure initiation is taken as the failure strength (Table 3).

Failure initiates at the weakest interface in the stacking sequence of a low-k specimen when the laser fluence reaches a critical value. The interfacial strengths of the failed interfaces for both Type 1-A and Type 2-A specimens are plotted in Fig. 11. The Si/OSG interface fails first for both untreated and He treated Type 1-A specimens at laser fluences 55 and 74 mJ/mm², respectively. The corresponding interfacial strengths at these critical fluences are 282 and 471 MPa (from Fig. 10(b)). The SiCN/OSG interface in untreated Type 2-A specimens failed at 63 mJ/mm² laser fluence with an interfacial strength of 372 MPa. In the case of He pretreated Type 2-A specimens, the OSG/Ti-Au interface debonded (at 71 mJ/mm²) before failure initiation in any of the low-k interfaces (Si/SiCN, SiCN/OSG). The adhesion strength of OSG/Ti-Au is 437 MPa (calibration curve not shown). Although we were unable to fail the low-k interfaces in the He pretreated Type 2-A specimens, a lower bound for the strength was estimated from the stress values at the critical laser fluence for failure of the OSG/Ti-Au interface. The maximum interface stress calculated at the OSG/SiCN interface was 453 MPa and the max stress at the Si/SiCN interface was 438 MPa. Hence, the strength of the OSG/SiCN interface was at least 453 MPa, denoted by ‘+++’ in the bar chart in Fig. 11.
Fig. 8. SEM images of spallation in low-κ specimens: (a) untreated Type 1-A specimen, (b) He pretreated Type 1-A specimen, and (c) Untreated Type 2-A specimen. The scanned image in panel (a) shows the specimen’s top view. The images in panels (b) and (c) are obtained by tilting specimens at 45° angle during scanning.

Fig. 9. Effects of laser fluence ($\Gamma$) and He pretreatment on the interfacial failure of Type 2-A specimens. Optical micrographs (a) and (b) correspond to the specimens which were not He pretreated whereas micrographs (c) and (d) show the spallation in He pretreated specimens.
4. Conclusions

The laser spallation technique was applied to induce failure at ultra thin multilayer low-κ (250 nm OSG and 30 nm SiCN) film interfaces. The interfacial strength of mutually switched stacked films on top of the Si substrate, fabricated in the presence and absence of He environment was evaluated. Based on a one dimensional wave propagation analysis, the test specimens were designed and prepared with a fs pulse shaping layer adhered to the back surface of the Si and an inertial Ti/Au layer deposited on the top surface of the low-κ specimen to enhance the interfacial stresses. Surface profilometry in combination with optical and SEM images was employed to identify the failed (weakest) interface in the stack. The adhesion strength of the failed interface was inferred from interferometric measurements performed on calibration (Si/SiN) samples and one-dimensional elastodynamic simulations of the multilayer specimen geometries. The Si/OSG and OSG/SiCN interfaces were characterized and the He pretreatment was found to significantly increase the interfacial strength for both specimen configurations. Failure could not be initiated at Si/SiCN interface due to experimental limitations and the high strength of this interface.

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