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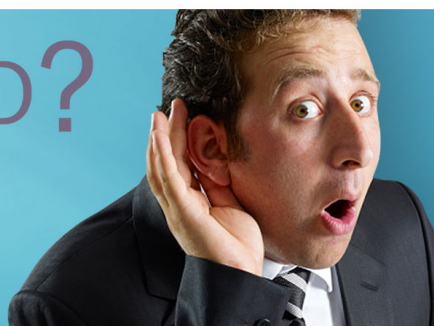
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## Selective interface toughness measurements of layered thin films

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Driven by the ongoing miniaturization and increasing integration in microelectronics devices, very thin metallic films became ever more important in recent years. Accordingly also the capability of determining specific physical and mechanical properties of such arrangements gained increasing importance. Miniaturized testing methods to evaluate, for example, the mechanical properties of thin metallic multilayers are therefore indispensable. A novel in-situ micromechanical approach is examined in the current study and compared to existing methods regarding their capability to determine the interface toughness of specific interfaces in multilayer configurations. Namely, sputter deposited copper and tungsten thin films with a thickness of approx. 500 nm on a stress-free silicon (100) substrate are investigated. The multilayer stacks consist of different materials that vary in microstructure, elastic properties and residual stress state. We examine the interface toughness via double cantilever beam tests, nanoindentation and novel miniaturized shear tests. The choice of a proper test method is indispensable when addressing strong interfaces, such as the W-Cu interface, in the presence of weaker ones. Finally, it is demonstrated that miniaturized shear testing is a very promising approach to test such strong interfaces as the interface of interest to fail is predefined by the sample geometry. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4978337>]

Over the past decades very thin metallic films became ever more important due to the ongoing miniaturization and increasing integration in microelectronic devices. Most often there are several thin films stacked on top of each other, thus interfaces between them are prominent sites of failure. As a consequence, it is of high interest to characterize the physical characteristics like the mechanical behavior and to quantify the critical energy release rate of the involved interfaces. As the film thickness for state-of-the-art microelectronics applications is frequently in the order of a few hundreds of nanometers, the spectrum of available micromechanical test methods is rather limited. A very fast and easy approach is to perform nanoindentation tests<sup>1,2</sup> (Figure 1a) to calculate the interfacial fracture energy by determining the buckle geometry around the indents. The limitation is that not all interfaces buckle upon indentation. Only compressively stressed interfaces are prone to delamination or a stressed overlayer has to be applied. As shown in Figure 1b, notched micro-cantilevers<sup>3</sup> can be employed to test the interface toughness of a layered material provided that the lever is long enough, meaning that the top layer must be rather thick. In the case of a layer stack with an individual layer thickness below 1  $\mu\text{m}$  this requirement is hardly fulfilled. A more promising approach is double cantilever beam (DCB) testing (Figure 1c),<sup>4</sup> where the interface of interest lies exactly in the middle of the specimen. The challenge is that both beams should have comparable elastic properties, so that a bending moment is applied symmetrically on the notched interface. With all described methods, it is more likely to test the weakest of the involved interfaces, which is not necessarily the one of interest. Therefore, a technique to test specific interfaces, even very strong ones in the presence of

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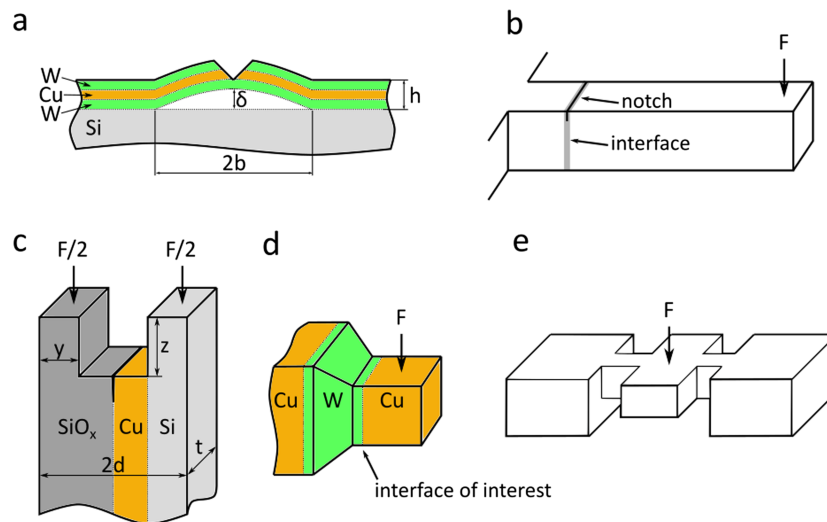


FIG. 1. Schematics of (a) the buckle geometry upon nanoindentation, (b) a notched micro-cantilever, (c) a DCB specimen, (d) a sample for shear testing of a specific interface (d) and (e) a double shear specimen.

weak ones, is of great importance. The idea is to perform a micro-shear experiment as depicted in Figure 1d, where the failing interface can be selected by the sample geometry. Therefore, the final shear specimen just contains the interface of interest. Thus, also strong material combinations can be tested by choosing the appropriate sample geometry even for very thin films. Although the idea of performing miniaturized shear tests is not new,<sup>5</sup> a double shear specimen similar to that shown in Figure 1e is only suitable for bulk materials and not for thin films.

In this work, different existing and novel approaches are examined regarding their capability to determine the interface toughness of specific interfaces in multilayer configurations. Specifically, we address copper (Cu) and tungsten (W) thin films deposited on a silicon (Si) substrate in various stack combinations. Using these materials, differing in microstructure and elastic properties, the strengths and weaknesses of the specific test methods concerning determination of the strength of the interfaces were able to be identified and are pointed out here. The investigated thin films were prepared via physical vapor deposition with an individual layer thickness of approx. 500 nm on a Si (100) substrate. Different configurations of the layer stacking such as W-Cu-W-Si and Cu-W-Cu-Si were tested. For more information concerning deposition parameters and microstructure the reader is referred to Ref. 6. On a second set of samples, an additional  $\text{SiO}_x$  film with a thickness of approx. 1.5  $\mu\text{m}$  was deposited. The  $\text{SiO}_x$  film was grown in a high vacuum deposition chamber by magnetically unbalanced reactive pulsed direct current magnetron sputtering. Deposition took place from a Si target in an Ar+O<sub>2</sub> reactive gas mixture at a total pressure  $p_{\text{TOT}} = p_{\text{Ar}} + p_{\text{O}_2} = 0.5 + 0.5$  Pa and a target current density  $I_d = 12$  mA/cm<sup>2</sup>. The substrate temperature was set to 200 °C and the pulsed substrate bias voltage (asymmetric bipolar pulse of 250 kHz) was kept constant at -100 V.

In sputter deposited films, either tensile or compressive residual stresses develop during fabrication, which may affect their mechanical behavior. Therefore, these stresses were assessed first by means of the ion beam layer removal (ILR) method.<sup>6-8</sup> The locally resolved residual stress profiles for each of the investigated stacks can be found in Ref. 6.

To access the toughness of the various interfaces different approaches were chosen. At first, the Cu-SiO<sub>x</sub> interface (Figure 1c) was tested via DCB experiments to find out if the interface is strong enough, so that the SiO<sub>x</sub> film could be used as a second beam to later quantify the toughness of the W-Cu interface in a DCB test. All micromechanical specimens in various geometries were prepared by focused ion beam (FIB) milling. Prior to FIB milling, the edge of the sample was polished using a Hitachi-E3500 Cross Section Polisher (Hitachi, Tokyo, Japan) to avoid any influence from breaking the coated wafer on the experimental results.<sup>9</sup> FIB milling was performed in a LEO 1540XB FIB workstation (Zeiss, Oberkochen, Germany) using gradually reduced ion beam currents for cutting

with finally 100 pA, and a notching current of 20 pA with a milling time per length of 40 s/ $\mu\text{m}$ . All DCB tests were performed in-situ in a LEO 982 SEM (Zeiss, Oberkochen, Germany) using an UNAT-SEM Indenter (Zwick GmbH & Co. KG, Ulm, Germany) equipped with a diamond flat punch (Synton-MDP AG, Nidau, Switzerland). The experiments were performed in displacement controlled mode with a loading speed of 10 nm/s. During each in-situ experiment, applied displacement and corresponding force were recorded and correlated with a video recorded at the same time. In Figure 2, representative force-displacement data of a DCB test of a  $\text{SiO}_x$ -Cu-Si specimen including screen shots throughout the test are depicted. Figure 2b shows the notched sample before the test, in Figure 2c the deflection of the crack into the  $\text{SiO}_x$ -Cu interface is evident and Figure 2d shows the completely fractured sample after the test. From the load-displacement curve in Figure 2a the maximum force at fracture  $F_c$  was obtained and the dimensions of the sample were measured from SEM images. The fracture resistance was then calculated using the following equation:<sup>4</sup>

$$G = 3F_c^2 \frac{(e - \mu z)^2}{Et^2 d^3}, \quad (1)$$

where  $\mu$  is the friction coefficient between punch and sample estimated as 0.1,<sup>10</sup> while  $e = (d-y)/2$  and  $d$ ,  $t$ ,  $y$  and  $z$  are the dimensions of the sample (see Figure 1c). According to Equation (1) the fracture resistance of the  $\text{SiO}_x$ -Cu interface is 16.3 J/m<sup>2</sup>.

Next, DCB specimens were prepared from a Cu-W-Cu-Si stack to test the W-Cu interface. For all tested samples the  $\text{SiO}_x$  film chipped off during the experiment, indicating that even though the highest load acted on the interface of interest, a geometrical less favored one failed first. Therefore, the interface toughness must be higher for the W-Cu than for the Cu- $\text{SiO}_x$  interface.

As a second approach to study the fracture resistance of the W-Cu interface, nanoindentation (Figure 1a) was chosen. The intention of the experiment was to induce opening the W-Cu interface by the presence of the W layer being highly compressively stressed.<sup>6</sup> The advantage of this nanoindentation test is that no other sample preparation is required except for a proper cleaning of the sample surface. The experiments were performed on a platform Nanoindenter G200 (Keysight Tec. Inc., Santa Rosa, CA, USA) equipped with a Berkovich and a spherical diamond indenter tip (Synton-MDP AG, Nidau, Switzerland). The tests with the Berkovich tip were conducted in a strain rate controlled mode with 0.05 s<sup>-1</sup> to a maximum indentation depth of 1  $\mu\text{m}$  including a superimposed CSM signal of 2 nm at 45 Hz. For the spherical tip with a radius of  $\sim 10 \mu\text{m}$  a standard load controlled operating scheme was applied to reach a maximum load of 100 mN after 30 s.

To analyze the resulting buckles a Veeco Dimension D3100 (Bruker Inc., Billerica, USA) atomic force microscope (AFM) was used in contact mode. The determination of the buckle geometry was done by analyzing the AFM images using Gwyddion<sup>11</sup> in order to collect the relevant information for calculating the adhesion energy. To check which interface actually delaminated, FIB cross-sections of the buckled indents were prepared. It was found that for all indents the W-Si interface buckled. From the AFM scans the buckle height  $\delta$  and the buckle width  $2b$  were determined. All equations to calculate the interfacial fracture energy were taken from Ref. 1 and 2. 8 buckles were measured and an interfacial fracture energy of  $48.3 \pm 13.3 \text{ J/m}^2$  for the W-Si interface was determined.

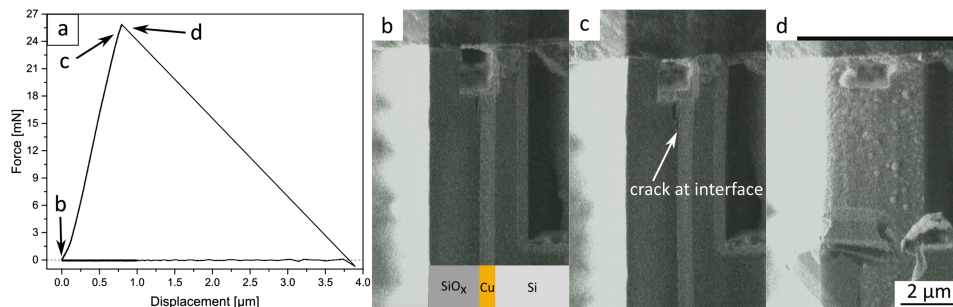


FIG. 2. Force-displacement data (a) and SEM-images of a  $\text{SiO}_x$ -Cu-Si DCB before the test (b), when the crack deflects into the interface (c) and after fracture (d).

With the results obtained to this point, it is evident that the W-Cu interface is the strongest one in this stack configuration. Therefore, as a third approach, specimens for a shear test on the W-Cu interface were prepared applying the same combination of ion slicing and FIB milling procedures as for the DCB samples. Shear specimens, as schematically shown in Figure 1d, were prepared in an Auriga FIB workstation (Zeiss, Oberkochen, Germany) with a current of 500 pA for coarse cutting and 20 pA for polishing. The final sample geometry is shown in Figure 3b. The displacement controlled tests were performed using a PI-85 Picoindenter (Hysitron Inc., Minneapolis, USA) with a conductive diamond flat punch (Synton-MDP AG, Nidau, Switzerland) at a loading speed of 4 nm/s. The force-displacement curve in combination with screenshots at specific points (a-e) is shown in Figure 3. In a first cycle, the crack initiated along the W-Cu interface (Figure 3c). In the subsequent cycle, the Cu top sheared off along the interface, after which the crack deflected into the W base of the sample (Figure 3d). To estimate the interface toughness, the work of separation is calculated from cycle 1. After Irwin's definition<sup>12</sup> the energy release rate is a measure of the available energy for a certain crack extension. Therefore, the area below the force-displacement curve after the first load drop, which corresponds to the available energy and is shown as grey hatched region in Figure 3a, is determined, and divided by the newly created fracture area. This calculation results in a value of approx. 230 J/m<sup>2</sup>. Thus, with this test an approximate interface toughness of the strongest W-Cu interface in the current layer stack was determined.

Comparing the interfacial fracture energy of the Cu-SiO<sub>x</sub> interface from the DCB tests to literature, we find that the values can range from 0.6 to 100 J/m<sup>2</sup>,<sup>13</sup> strongly depending on the film thickness that spanned from 40 to 3000 nm. Therefore, the value of 16 J/m<sup>2</sup> for a 500 nm film obtained in the current study is well in the range of the literature data.

Putting the three test methods and the gathered data into context, it becomes evident why the evaluation of fracture resistance of the W-Cu interface with either DCB or indentation test methods is not possible. For the DCB test, although the W-Cu interface is subjected to the highest load, the other interfaces are much weaker and consequently fail well before the interface of interest. The same holds true for the indentation experiments, where the fracture also occurs preferentially along the weakest interface, which is additionally promoted by the present residual stress state. The characterization of strong interfaces, especially in layered systems, where constituents of various interface fracture resistance are combined, is rather challenging. Nevertheless, by shear testing the W-Cu interface can be addressed. A relatively high value of 230 J/m<sup>2</sup> is derived for several reasons. At first, only a mode II behavior can be studied by a shear test, whereas for the DCB test and nanoindentation a mode I failure is tested, being far more critical. Furthermore, the soft Cu undergoes plastic deformation

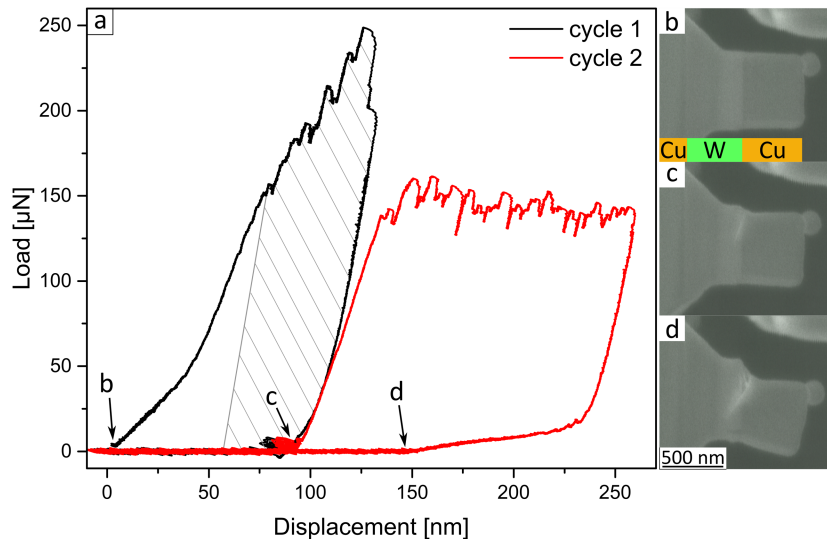


FIG. 3. Force-displacement data (a) of a shear test on a W-Cu interface and SEM-images of the sample before the test (b), between cycle 1 and 2 (c) and after the test (d).

in contact with the flat punch indenter. This is evidenced by the shape of the initial contact of the first loading curve (Figure 3a), where the slope of the curve changes from a lower to a higher value hence full contact between sample and flat punch indenter is achieved at approx. 50  $\mu\text{N}$ . The value of 230  $\text{J/m}^2$  for a mixed mode interface toughness of the W-Cu interface with a dominant mode II component is thus an upper limit rather than an accurate value. Additionally, the effect of residual stresses on the interfacial fracture energy is not evaluated. It was already demonstrated in Ref. 8 that the sample geometry influences the residual stress state and that the stresses relax for a decreased sample size. Therefore, the small samples investigated by shear testing facilitate stress relaxation and residual stresses can be neglected as the intrinsic interface toughness is examined. Nevertheless, it can be expected that for nanoindentation and the DCB test the initial residual stresses still play a role concerning the interface toughness, which is not quantified in the current study.

In conclusion, the choice of proper method to determine an interface toughness value is very crucial and strongly depends on the interface and material of interest. Especially in the case of very thin films, many methods such as notched micro-cantilever bending and double shear specimens are not applicable due to physical material limits. In this work, it was shown that even for very thin films there are methods to test strong interfaces in the presence of weak ones by utilizing a novel shear testing approach as sample geometry. Applying shear testing is very promising, as in a multilayer system the failing interface can simply be predefined by the sample geometry.

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- <sup>1</sup> M. Kriese, W. Gerberich, and N. Moody, "Quantitative adhesion measures of multilayer films: Part I. Indentation mechanics," *J. Mater. Res.* **14**(7), 3007–3018 (1999).
- <sup>2</sup> M. Cordill, D. Bahr, N. Moody, and W. Gerberich, "Recent developments in thin film adhesion measurement," *IEEE Transactions on Device and Materials Reliability* **4**(2), 163–167 (2004).
- <sup>3</sup> K. Matoy, T. Detzel, M. Müller, C. Motz, and G. Dehm, "Interface fracture properties of thin films studied by using the micro-cantilever deflection technique," *Surf. Coat. Technol.* **204**, 878–881 (2009).
- <sup>4</sup> S. Liu, J. Wheeler, P. Howie, X. Zeng, J. Michler, and W. Clegg, "Measuring the fracture resistance of hard coatings," *App. Phys. Lett.* **102**, 171907 (2013).
- <sup>5</sup> J.-K. Heyer, S. Brinckmann, J. Pfetzinger-Micklich, and G. Eggeler, "Microshear deformation of gold single crystals," *Acta Mater.* **62**, 225–238 (2014).
- <sup>6</sup> R. Tremel, D. Kozic, J. Zechner, X. Maeder, B. Sartory, H.-P. Gänser, R. Schöngrundner, R. Brunner, and D. Kiener, "Local determination of residual stress gradients in single- and multilayer thin film systems with high resolution," *Acta Mater.* **103**, 616–623 (2016).
- <sup>7</sup> S. Massl, J. Keckes, and R. Pippan, "A direct method of determining complex depth profiles of residual stresses in thin films on a nanoscale," *Acta Mater.* **55**, 4835–4844 (2007).
- <sup>8</sup> R. Schöngrundner, R. Tremel, T. Antretter, D. Kozic, W. Ecker, D. Kiener, and R. Brunner, "Critical assessment of the determination of residual stress profiles in thin films by means of the ion beam layer removal method," *Thin Solid Films* **564**, 321–330 (2014).
- <sup>9</sup> S. Wurster, R. Tremel, R. Fritz, M. W. Kapp, E. Langs, M. Alfreider, C. Ruhs, P. J. Imrich, G. Felber, and D. Kiener, "Novel methods for the site specific preparation of micromechanical structures," *Practical Metallography* **52**, 131–146 (2015).
- <sup>10</sup> F. Bowden and A. Hanwell, "The friction of clean crystal surfaces," *Proc. Roy. Soc. A* **295**, 233–243 (1966).
- <sup>11</sup> D. Nečas and P. Klapetek, "Gwyddion: An open-source software for SPM data analysis," *Central European Journal of Physics* **10**(1), 181–188 (2012).
- <sup>12</sup> G. Irwin, "Onset of fast crack propagation in high strength steel and aluminum alloys," in *Sagamore Research Conference Proceedings*, Vol. 2 (1956), 289–305.
- <sup>13</sup> A. Volinsky, N. Moody, and W. Gerberich, "Interfacial toughness measurements for thin films on substrates," *Acta Mater.* **50**, 441–466 (2002).