Significance of Adhesion and Interfacial Fracture in Design of Reliable Nanomaterials and Nanodevices

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NIST

Workshop on Reliability Issues in Nanomaterials

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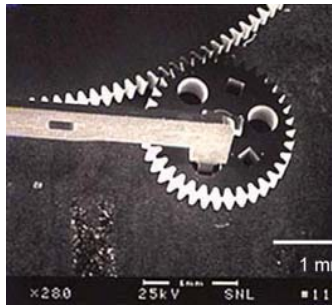
Our interests encompass many activities where the interfaces control performance and reliability.

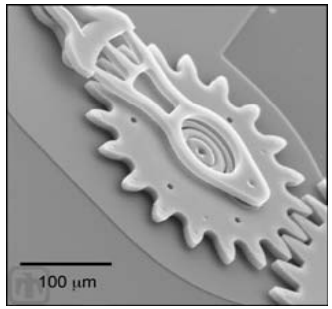
- Structural materials where exposure to active environments can lead to premature failure along interfaces.
- Welds where defects, elemental segregation, and second phase precipitation can sensitize interfaces to fracture.
- Thin films where the composition and structure of the films controls resistance to wear, corrosion and the performance and reliability of thin film microelectronic devices.

Adhesion and fracture of interfaces is critical to performance and reliability of nanostructured materials and devices.

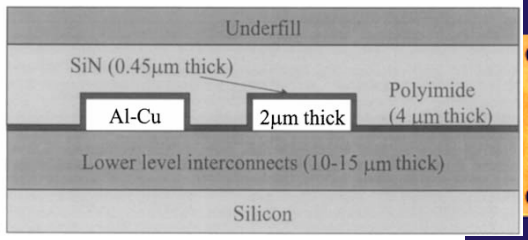
Many Sandia programs incorporate thin films and nanostructured materials where performance and reliability must be assured.

LIGA

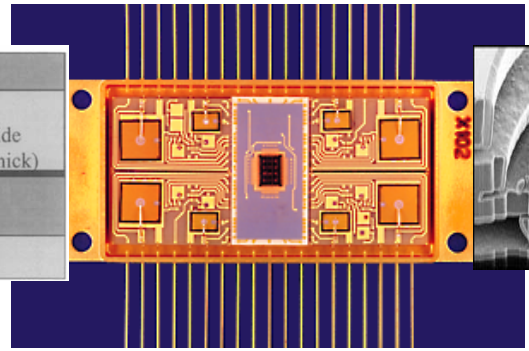


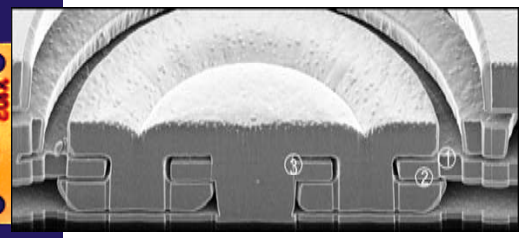


MEMS

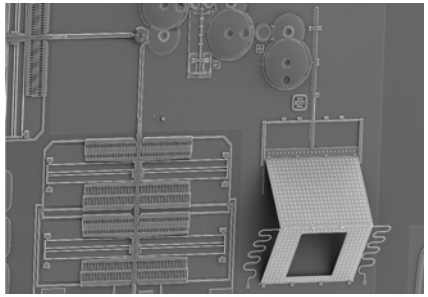


microelectronics

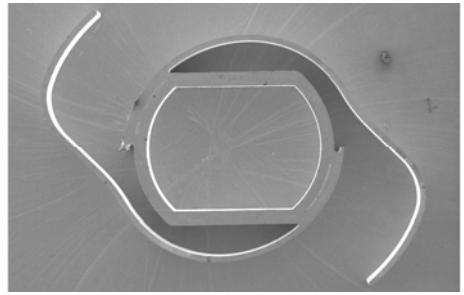




protective coatings



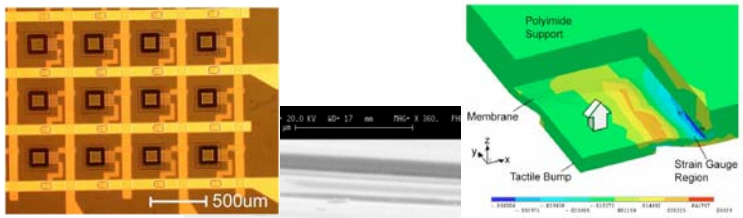
MEMS mirrors

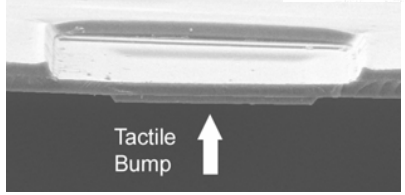


Micro springs

Thin films and nanostructured materials are the basis for emerging technologies that will dominate near term advances in nanotechnology.

MEMS Based Tactile Sensor



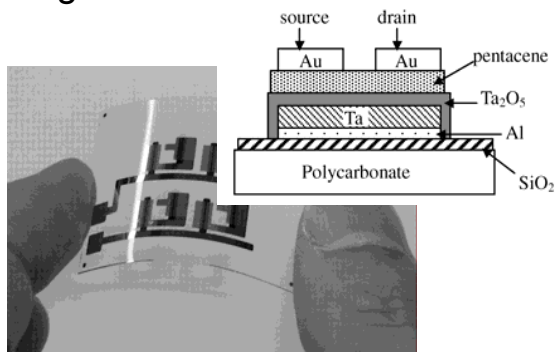


(Engle 2003)

Sensate Liner

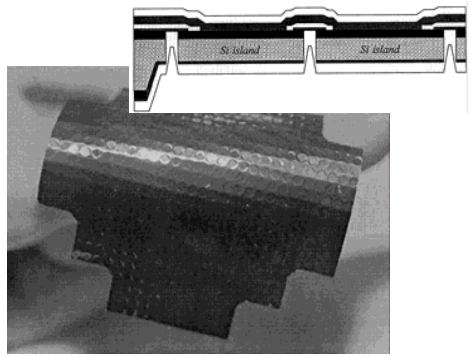
(Cadogan 2003)

Organic Thin Film Transistor



(Inoue 2003)

Flexible skin



(Jiang 2000)

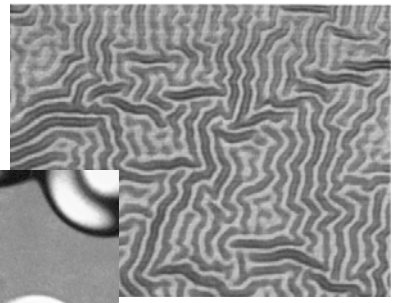
These are first generation devices where limited effort has been devoted to the underlying science of adhesion.

Thin films of metals, ceramics, and polymers are typically subject to appreciable residual stress that can trigger deformation and fracture.

Compressive stresses can lead to wrinkling, delamination, and buckling.

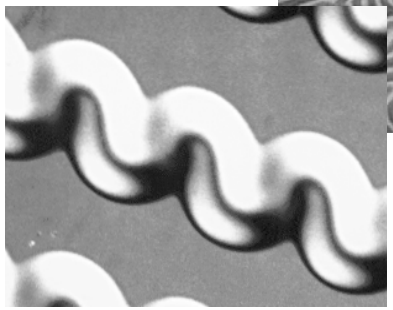
Tensile stresses can lead to film fracture and decohesion.

wrinkling



(Lacour 2003)

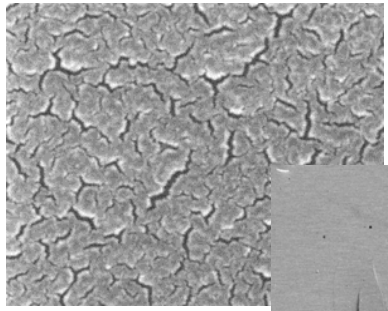
buckling



(Moody 2001)

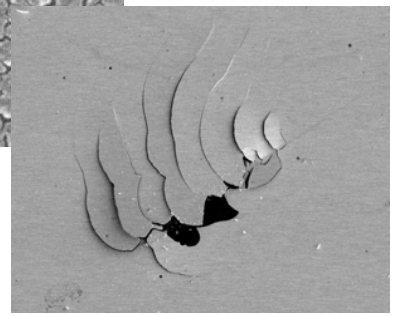
delamination

mudflat cracking



(Lacour 2003)

channel cracking



(Moody 2004)

decohesion

Adhesion and fracture are critical issues

Questions for Characterization and Testing

Which property measurements are critical for design of reliable nanomaterials and devices?

Are existing test methods sufficient?

What is the role of modeling?

What answers does modeling provide?

Experimental and theoretical approaches must be combined for quantitatively correct properties and physically accurate processes.

Questions will be addressed using specific examples

Test methods and property characterization techniques

Deformation and fracture of soft gold films (microelectronics, MEMS)

- significance
- properties
- mechanisms

Fracture of hard ALD tungsten films (nanodevices)

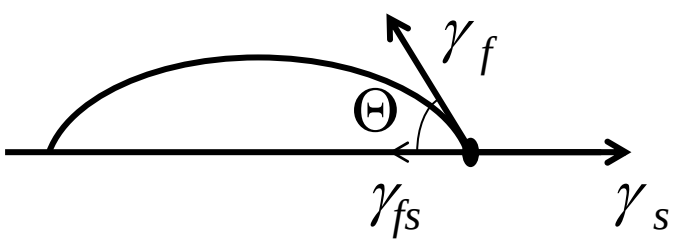
- significance
- properties
- mechanisms

Adhesion is defined as the total irreversible energy per unit area required to separate materials at the interface.

True work of adhesion is the energy required to create free surfaces

$$W_A = \gamma_f + \gamma_s - \gamma_{fs}$$

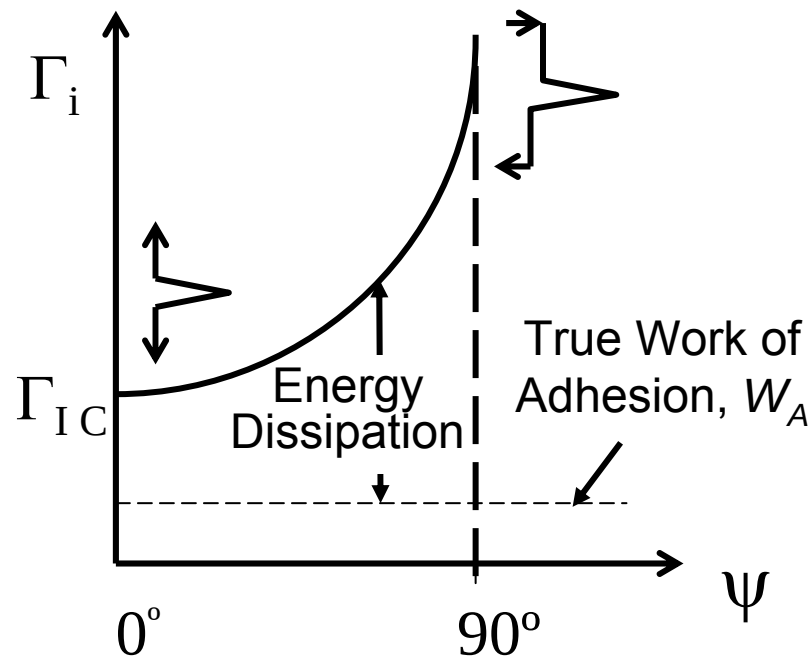
It is often determined by contact angle measurements



$$\gamma_{fs} = \gamma_s - \gamma_f \cos\Theta$$

$$W_A = \gamma_f + \gamma_s - \gamma_{fs} = \gamma_f(1 - \cos\Theta)$$

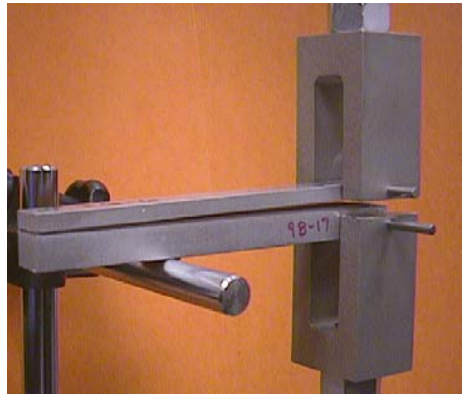
Practical work of adhesion measured by delamination

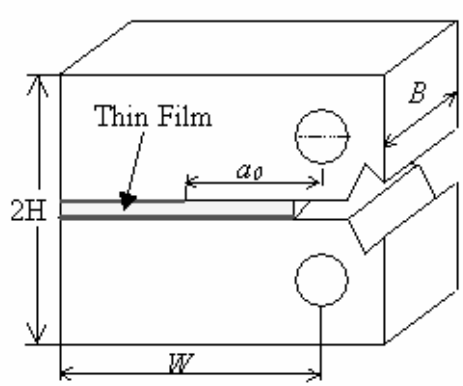


Often includes energies from additional processes

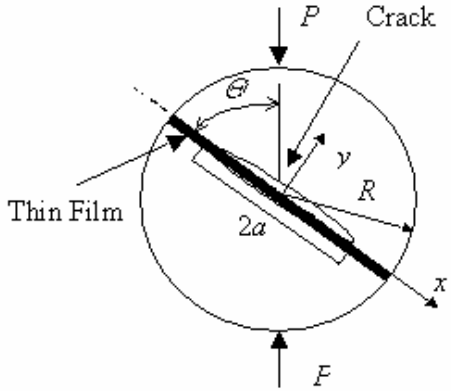
Traditional Test Techniques

DCB Sample

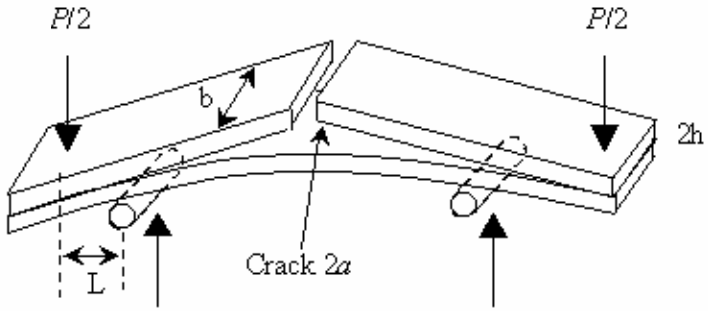




K_{Ic} CTS



Brazil Nut Sample



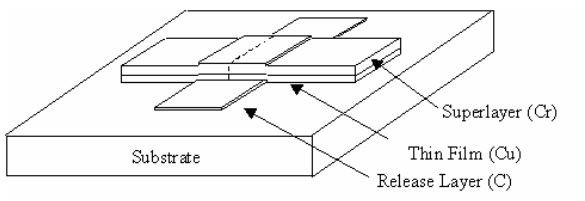
Four Point Bend Sample

Material Intensive Tests

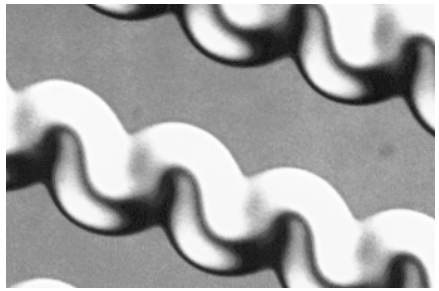
Evolving Microscale Tests

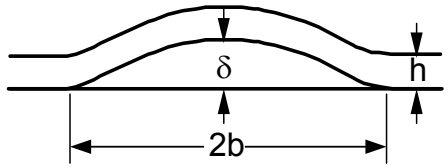
Stressed overlayer and nanoindentation test techniques

Stressed Overlayer

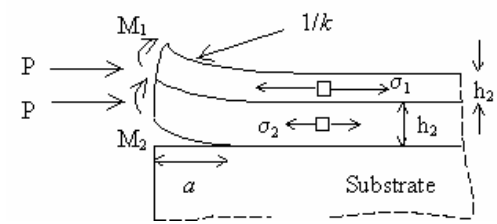


uniform width blisters

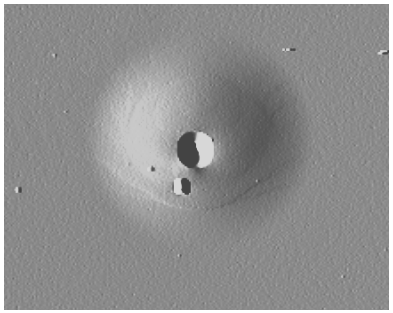


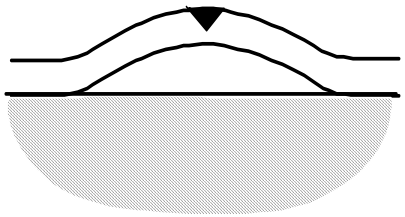


Edge Lift Off Test

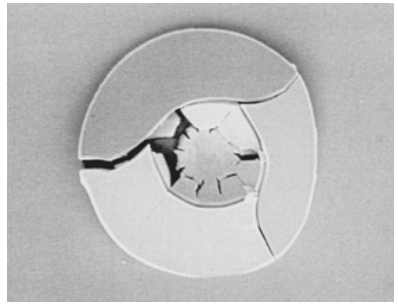


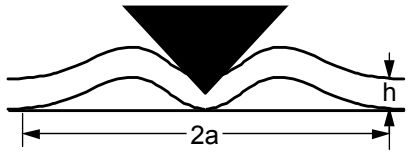
single buckle blisters





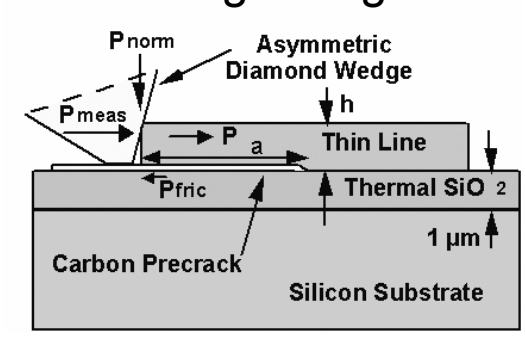
double buckle blisters



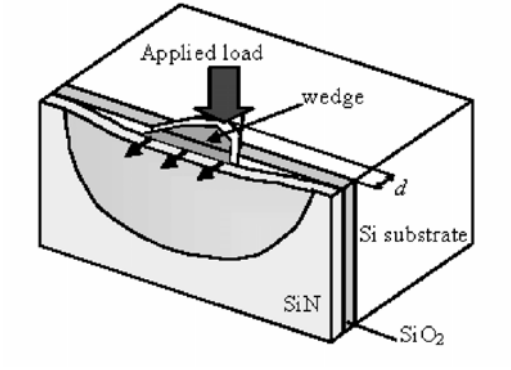


Nanoscratch Test Techniques

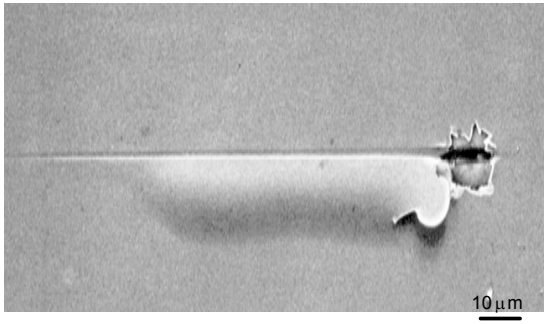
Sliding Wedge

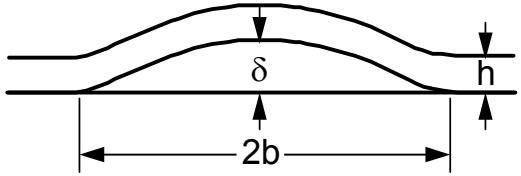


Cross-Sectional Scratch

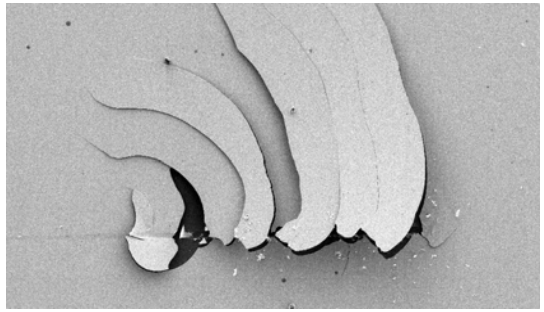


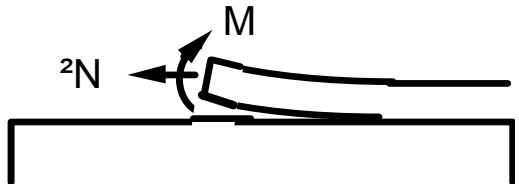
uniform width blisters



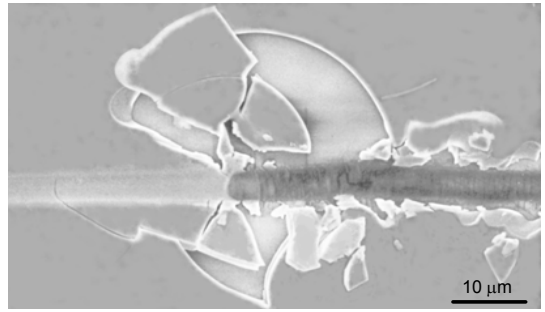


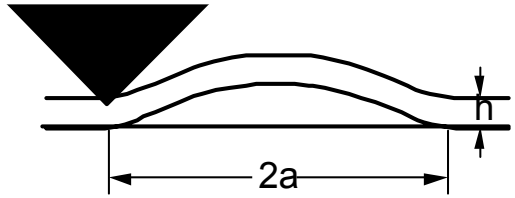
Channel cracking and decohesion





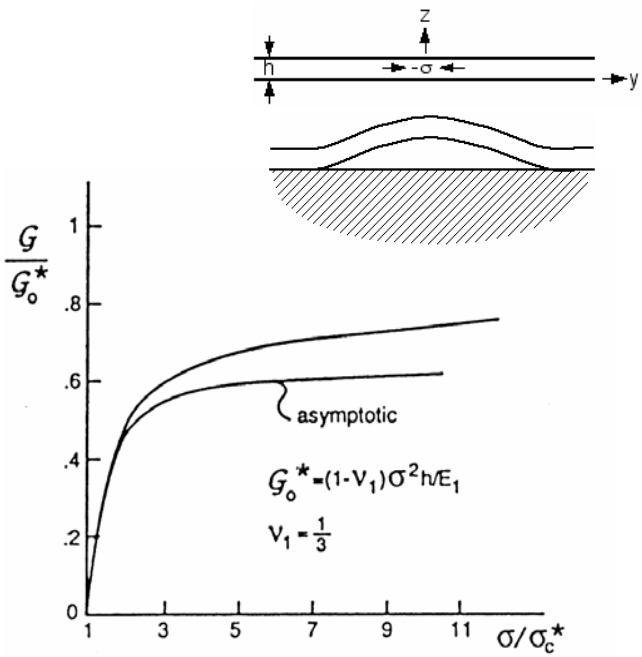
single buckle blisters



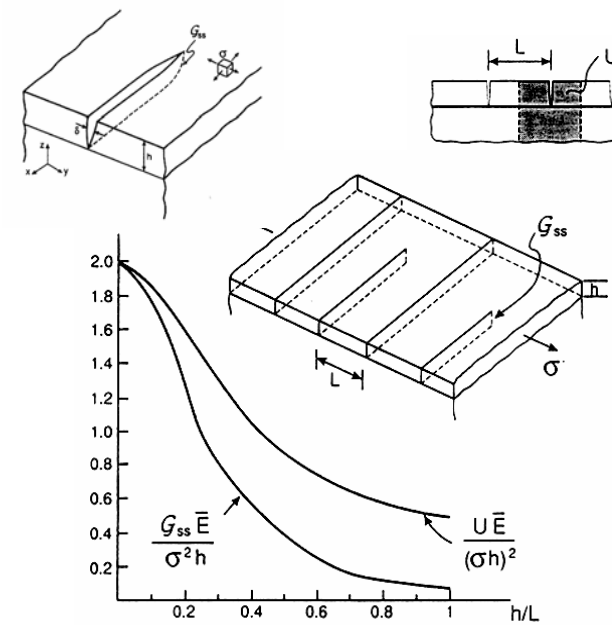


There is a large body of elastic thin film on rigid substrate and elastic contact mechanics solutions describing fracture

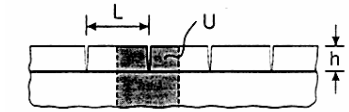
Delamination and Buckling



Single Crack



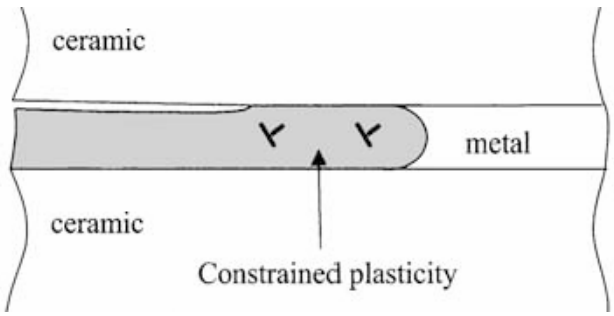
Multiple Cracks

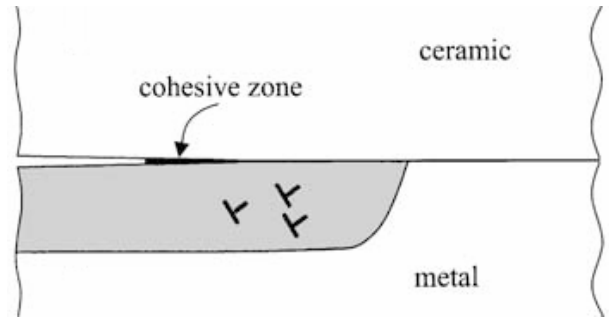


Hutchinson and Suo (1992); Hu, Thouless, Evans (1988); Thouless (1990; Ye, Suo, Evans (1992); Beuth (1992)

This is a very difficult problem leading to solutions based on simple geometric idealizations and the concept of steady state cracking.

The introduction of plasticity, especially substrate plasticity, significantly complicates the problem.





(Suo 2003)

In compression, it increases the energy for decohesion
In tension, it further drives cracking and delamination

- It is difficult to perform suitable mechanical tests.
- It is difficult to deconvolute film, substrate and interface contributions.

- Conventional continuum plasticity theories lack an internal length scale.

Nicola, Van der Giessen, Needleman 2003

As a result, the critical relationships between adhesion, properties, and fracture are not well defined.

Tests and models are based on idealized elastic systems

Analytical modeling and elastic concepts can define fracture in select cases

Advances are incremental through modification of existing tests and models

As films get thinner and structures get smaller, modifications become problematic

Advances are especially difficult for viscoelastic and plastically deforming systems

Deformation and Fracture of Thin Gold Films

M. Cordill, D. P. Adams, D. Medlin, N. Yang, D. F. Bahr, A. A. Volinsky, W. W. Gerberich,

Which property measurements are critical for design of reliable nanomaterials and devices?

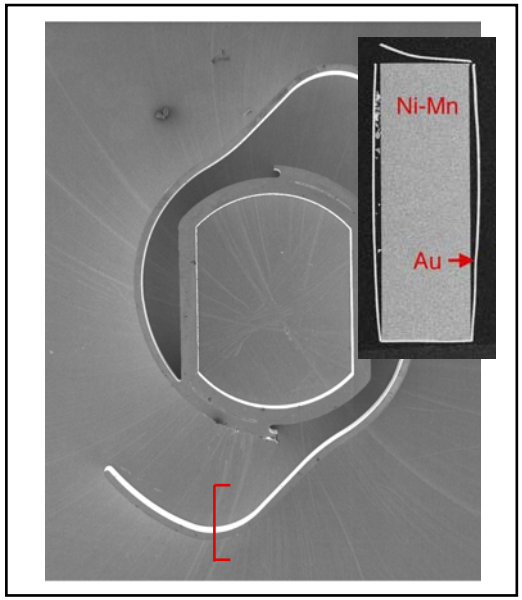
Are existing test methods sufficient?

What is the role of modeling?

What answers does modeling provide?

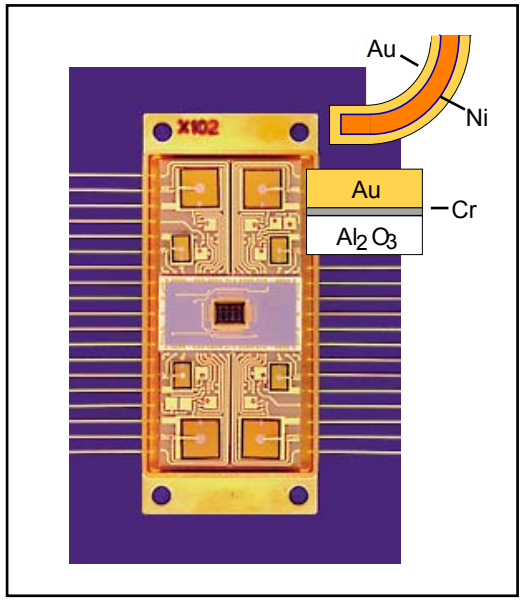
Thin gold films are used in many Sandia designed components from microelectronics and MEMS mirrors to LIGA springs.

LIGA Springs



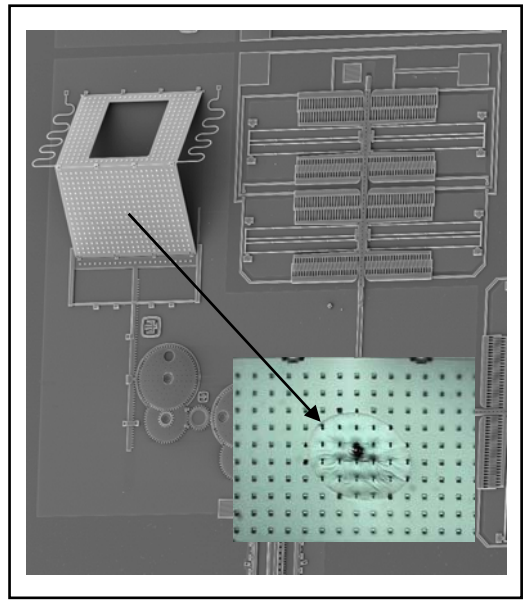
Processing

Microelectronics



Aging

MEMS Mirrors

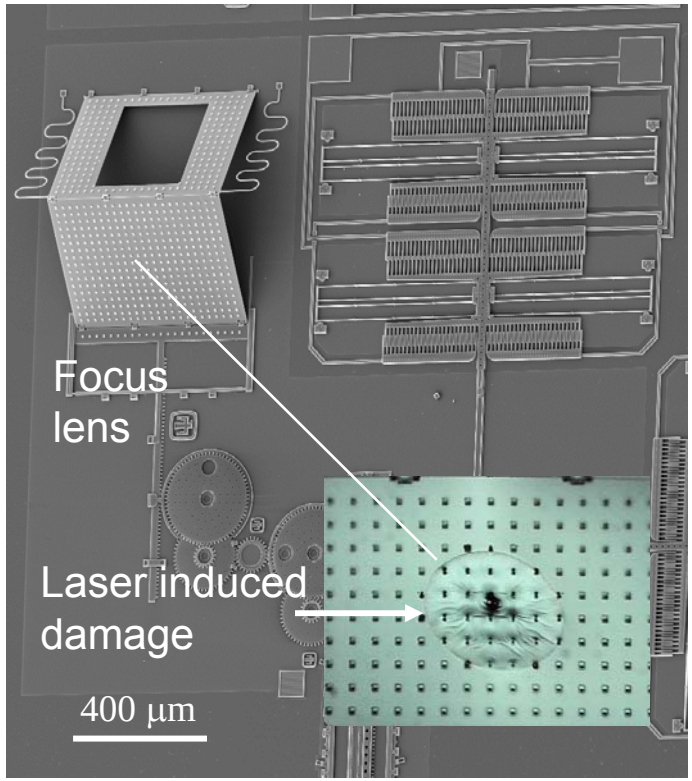


Service

Gold film adhesion is critical to performance and reliability

Heat absorption leads to surface damage and mirror failure in silicon MEMS mirrors

MEMS mirror system with comb-drive actuators



(J.J.Sniegowski, M.DeBoer, 2000)

Wavelengths of Interest:
Infrared 890 nm
(Externally powered MEMS)

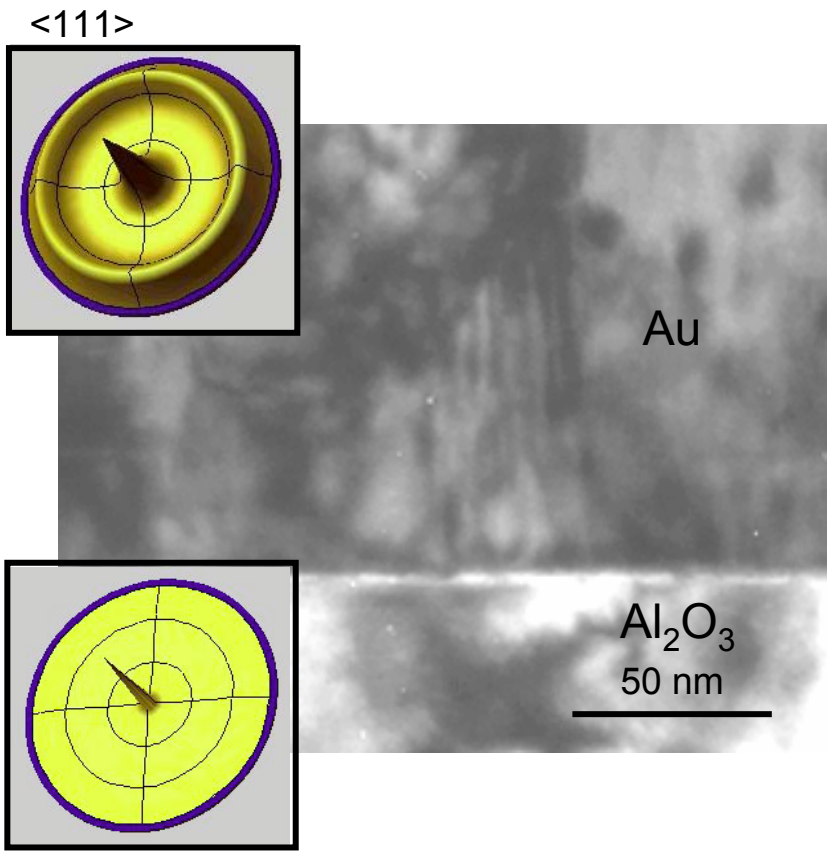
Mirror dimensions
Material : Polycrystalline Si
Si mirror thickness: 2.4 µm
Diameters: 100 µm – 2 mm

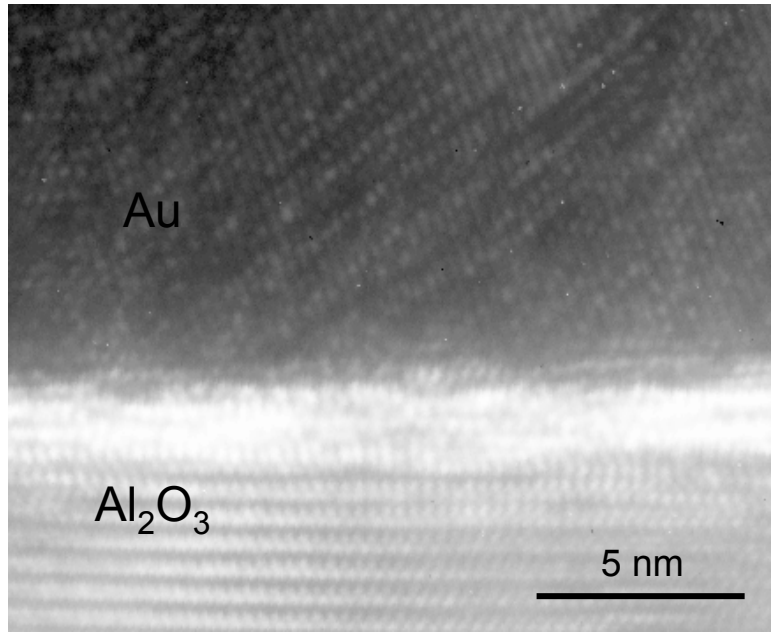
Coefficient of reflection for Si is less than 0.3

(D. P. Adams, Y. N. Picard, O. B. Spahn, S. M. Yalisove, J. Sobczak, K. Archuleta, V. C. Hodges, 2002)

Gold films maximize reflection and minimize film stress and heat absorption

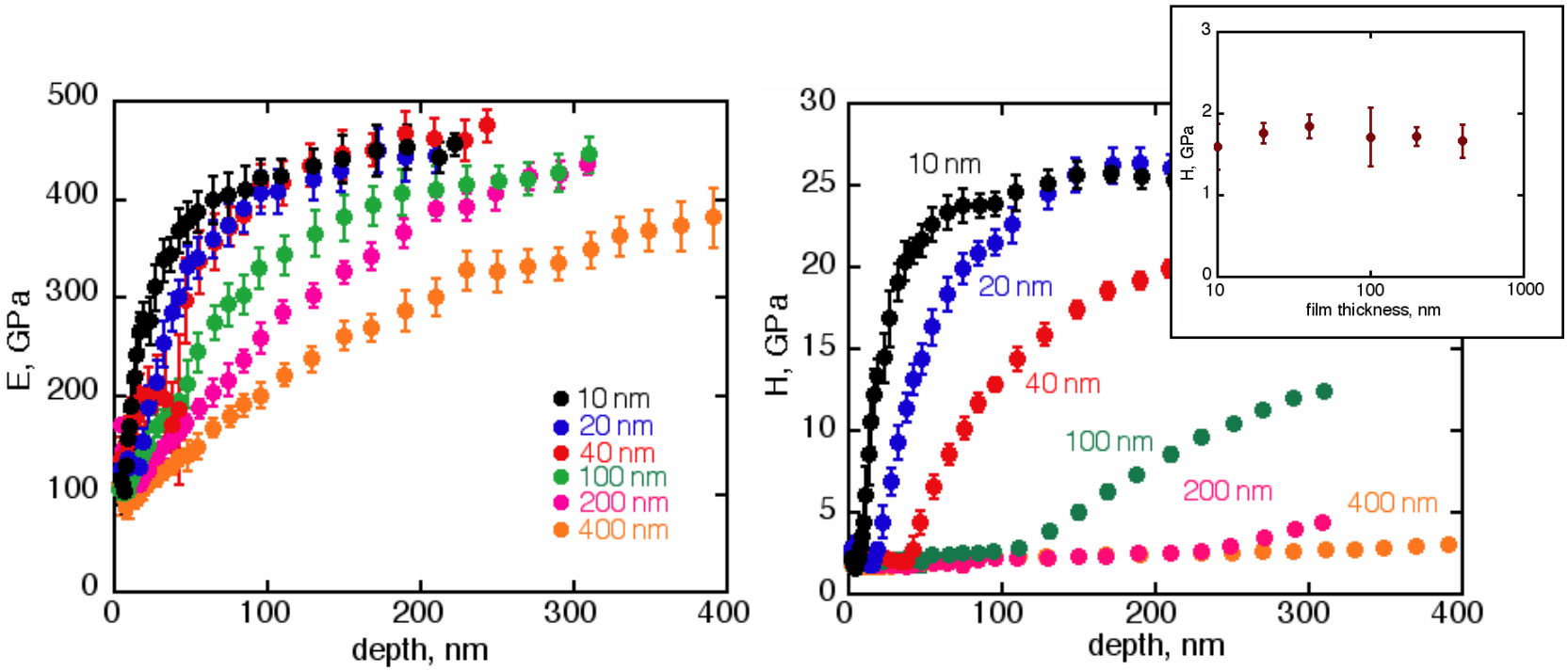
Studied effects of film thickness adhesion of sputter deposited gold films on (0001) sapphire substrates.





These films went down with a random in-plane texture and no registry across the interface.

Elastic and plastic properties are needed for assessment of film performance.



The elastic modulus is consistent with a [111] normal film. The high hardness results from a 20 nm grain size.

Continuous stiffness techniques enable large scale data acquisition

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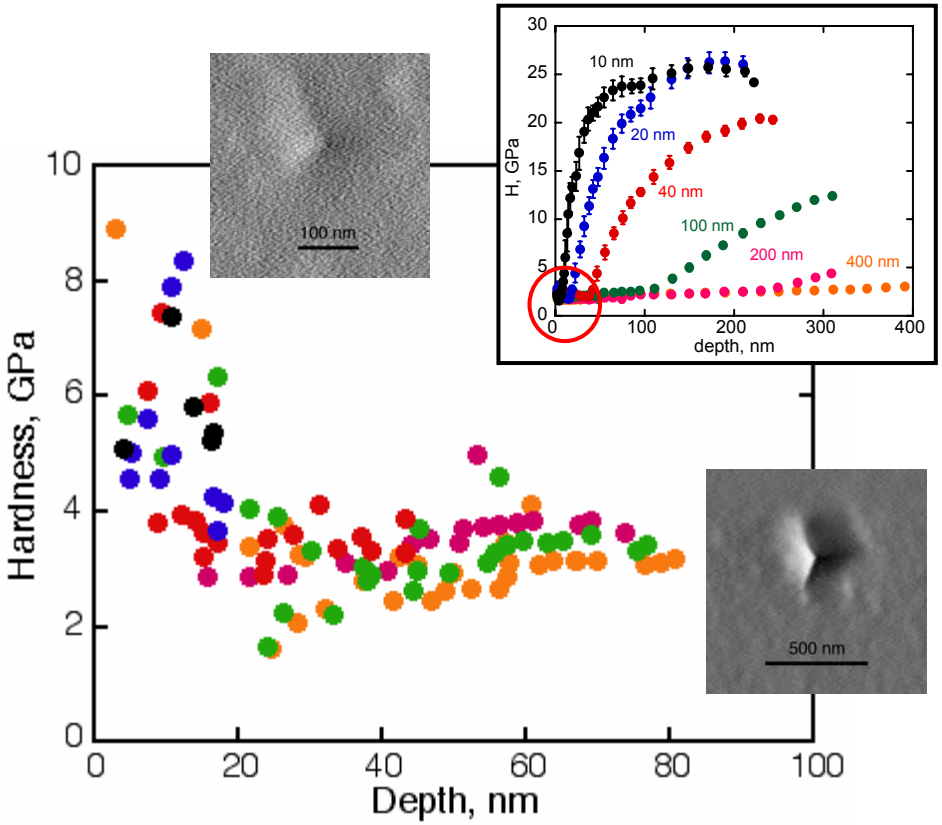
These techniques are based on tip shape functions for calculation of contact depths and contact areas

- never fully describe contact
- do not accurately reflect deformation processes

Understanding requires direct imaging of contact

AFM based indentation measurements show a strong indentation size effect on the measured hardness of thin gold films



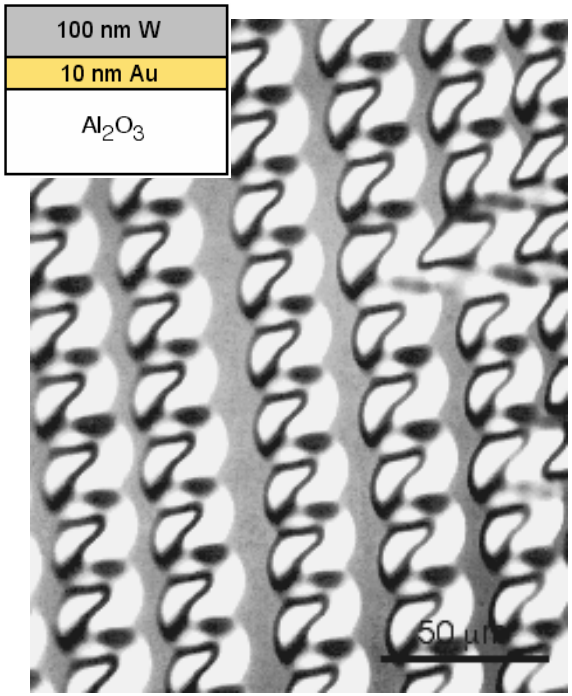
(M. J. Cordill)

- Probe radius exceeds feature (grain/film) size
- Pile-up and sink-in markedly require direct measurement

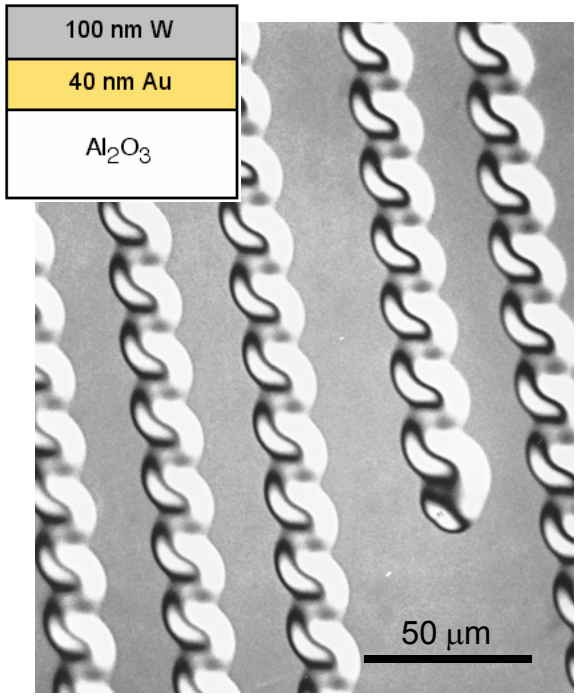
Material behavior at very small scales dominates deformation behavior

Tungsten overlayers triggered extensive delamination and telephone cord blister formation in all films.

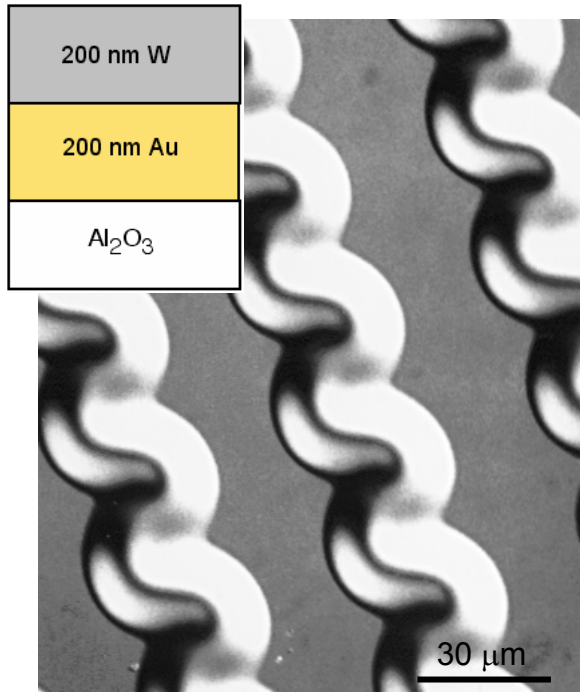
10 nm Au



40 nm Au

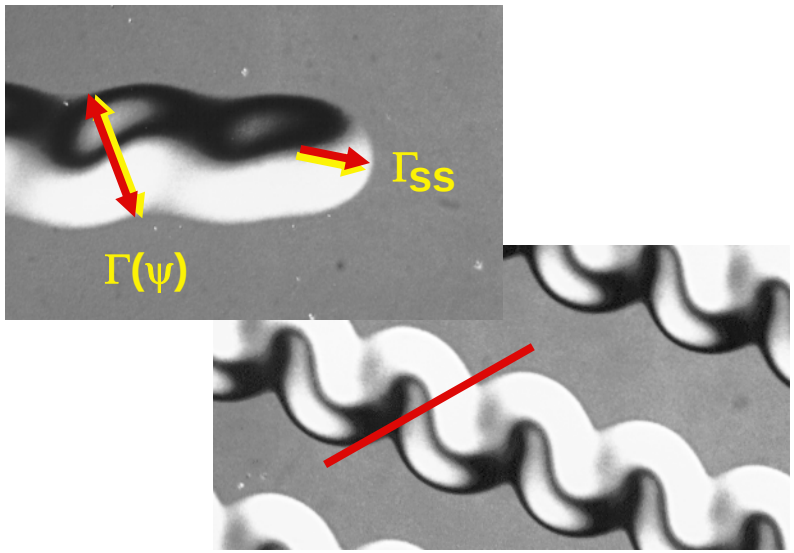


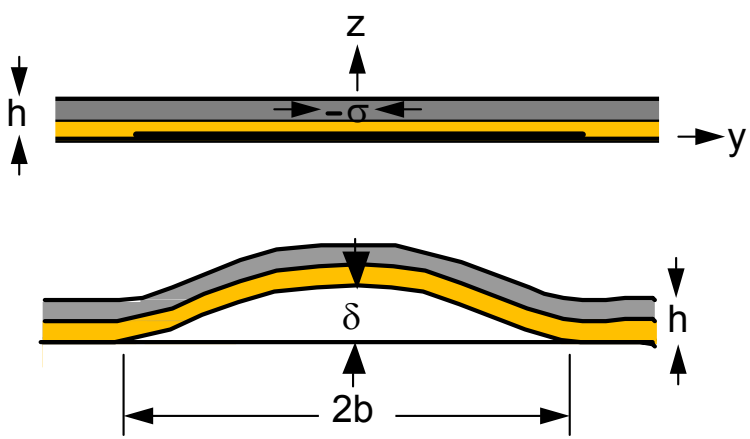
200 nm Au



Thick films exhibited a greater apparent resistance to delamination and buckling.

Mechanics-based modeling gives us the stresses and fracture energies for film failure of compressively stressed films





In terms of a single set of parameters, the stress for delamination is,

$$\bar{\sigma}_b = \frac{\mu^2}{Bhb^2}\left[\frac{E_1}{\left(1-\nu_1^2\right)}\right]\left(I_T\right)$$

Residual stress is as follows,

$$\bar{\sigma}_r = \bar{\sigma}_b\left[\frac{3}{4}\left(\frac{\delta^2}{h^2}+1\right)\right]$$

The strain energy released along the side wall is given by,

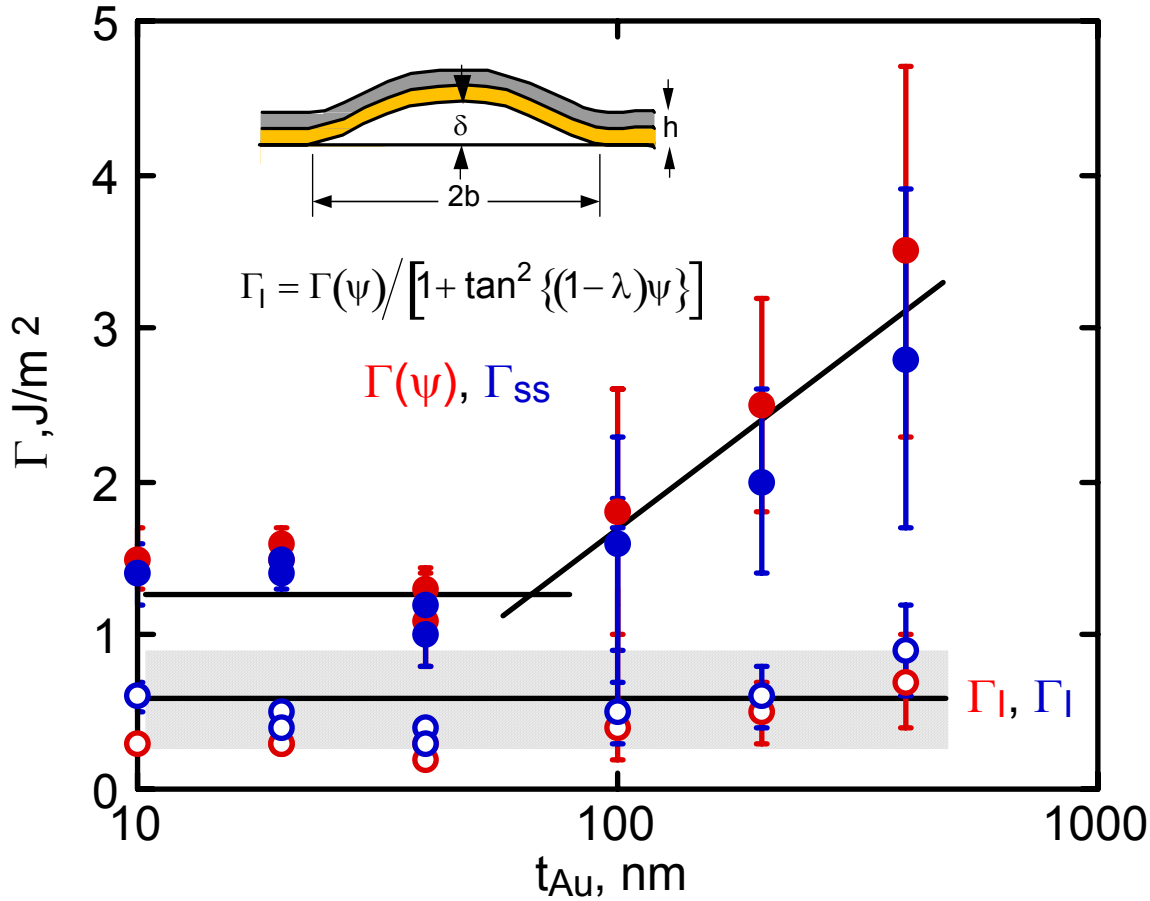
$$\Gamma(\psi) = \left[\frac{\left(1-\bar{\nu}^2\right)h}{2\bar{E}}\right]\left(\bar{\sigma}_r-\bar{\sigma}_b\right)\left(\bar{\sigma}_r+3\bar{\sigma}_b\right)$$

and along the propagating curved front.

$$\Gamma_{SS} = \left[\frac{\left(1-\bar{\nu}^2\right)h\bar{\sigma}_r^2}{2\bar{E}}\right]\left(1-\frac{\bar{\sigma}_b}{\bar{\sigma}_r}\right)^2$$

Hutchinson and Suo (1992)
Bagchi and Evans (1992)
Kriese, Moody, Gerberich (1998)

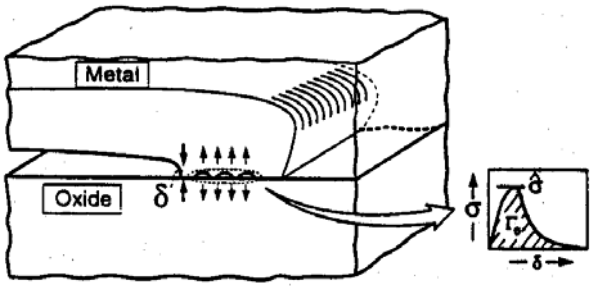
The mode I component is independent of film thickness and equal to the true work of adhesion for gold on sapphire.



Adhesion controls susceptibility to interfacial fracture in very thin films

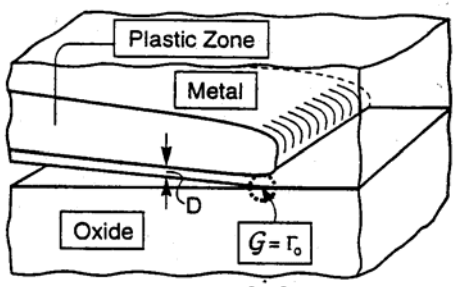
Modeling is particularly valuable for defining the role of fracture processes on film fracture

crack tip blunting



plastic zone EPZ model

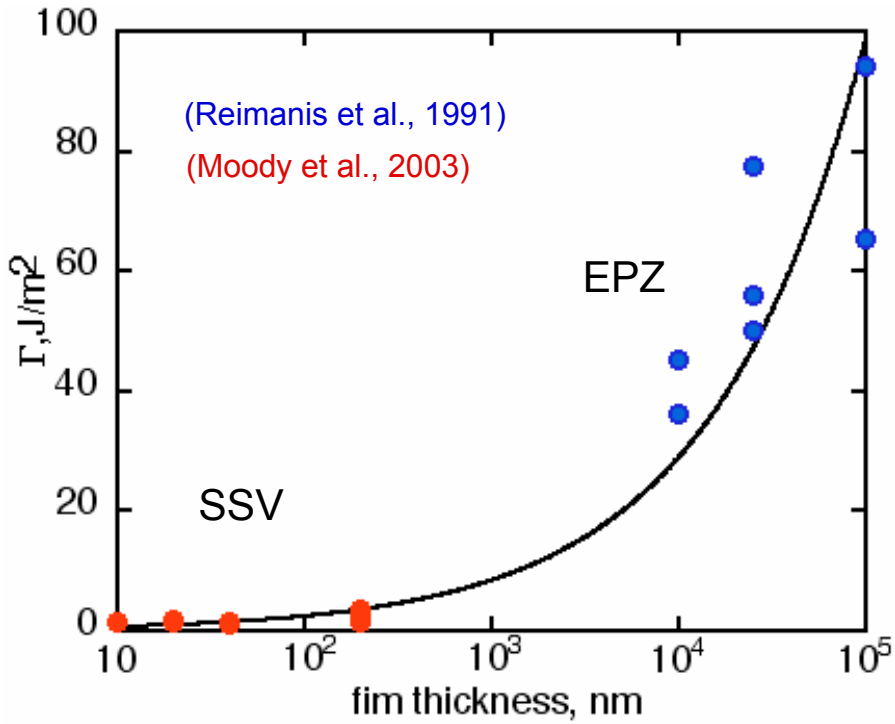
atomically sharp crack



elastic zone SSV model

(Evans, Hutchinson, Wei, 1999)
(Suo, Shih, Varias, 1993)

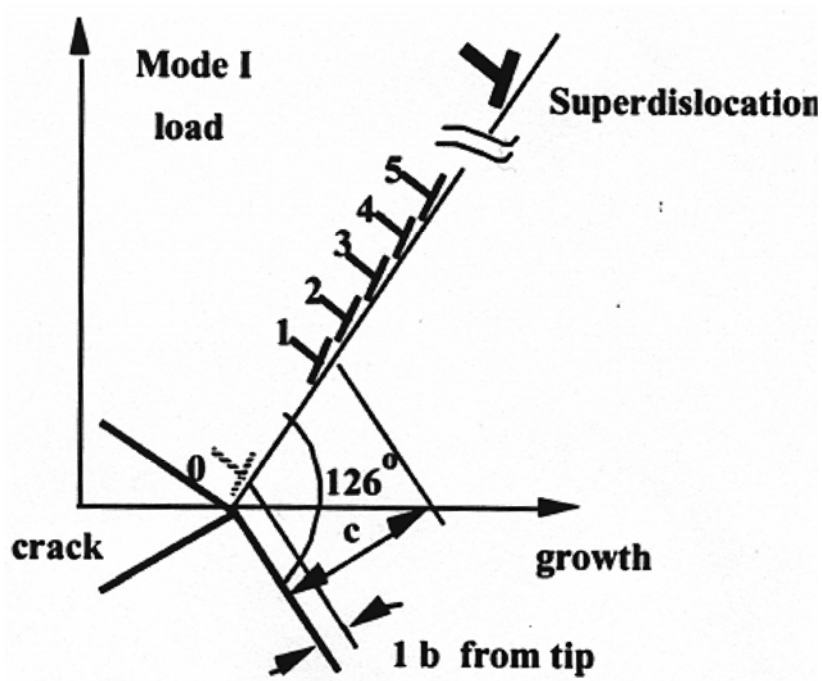
Gold Films



ductile to brittle transition

No analytical model effectively integrates elastic and plastic processes

Modeling dislocation behavior at a crack tip using a discrete dislocation approach provides a physical basis for elastic fracture.



Dislocation shielding creates a dislocation free zone

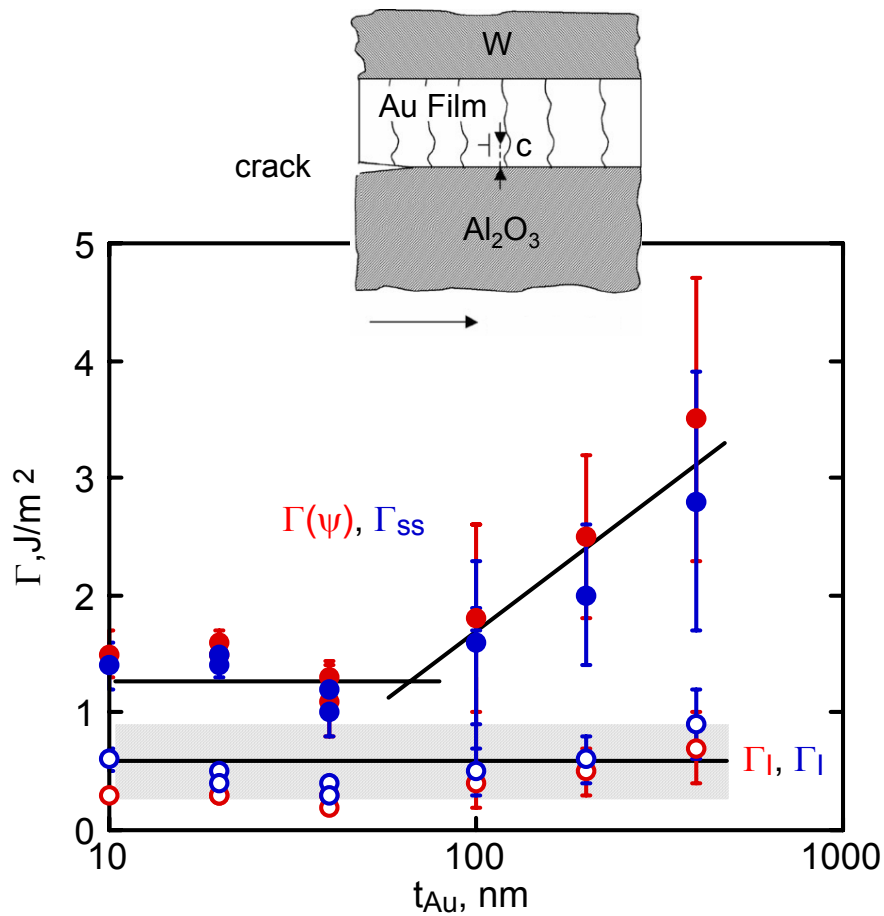
$$k_I = \frac{3}{\pi}\sqrt{\frac{2}{\pi}}\sigma_{ys}\sqrt{c}\left[\ln\left(\frac{4R_p}{c}\right)+\frac{4}{3}\right]$$

k_I = crack tip normal stress intensity
R_p = plastic zone size
c = dislocation free zone

(Thomson 1986)
(Lin,Thomson 1986)

Competition between applied stresses and image forces creates a DFZ

Formation of a dislocation free zone can explain elastic interfacial fracture in thin gold films



Conditions for fracture are given by,

$$k_I = \frac{3}{\pi}\sqrt{\frac{2}{\pi}}\sigma_{ys}\sqrt{c}\left[\ln\left(\frac{4R_p}{c}\right)+\frac{4}{3}\right]$$

k_I = crack tip normal stress intensity
R_p = plastic zone size
c = dislocation free zone

At the transition, h~ 90 nm :

k_I = 0.22 MPa$\sqrt{m}$
$c = R_p$ = 10-30 nm

(Thomson 1986)
(Lin,Thomson 1986)

High dislocation densities can exist in nc-materials

Do crack tip dislocations interact with exisitng dislocations?

Do we have separate independent dislocation populations?

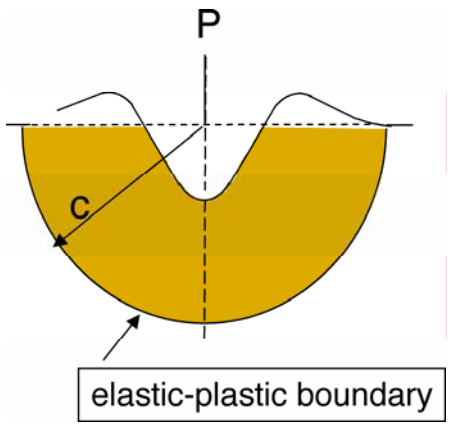
Do we really have a dislocation free zone?

Do we need to address nucleation and propagation?

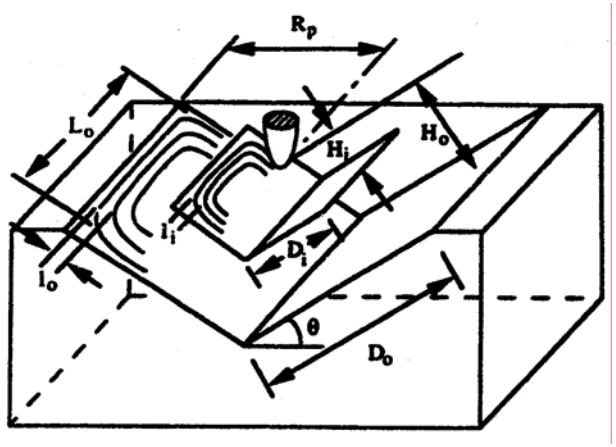
Direct observation can alter stress state and dislocation structure

A potential solution employs AFM-based nanoindentation to simulate a crack

Plastic zone formation



Indentation induced dislocations

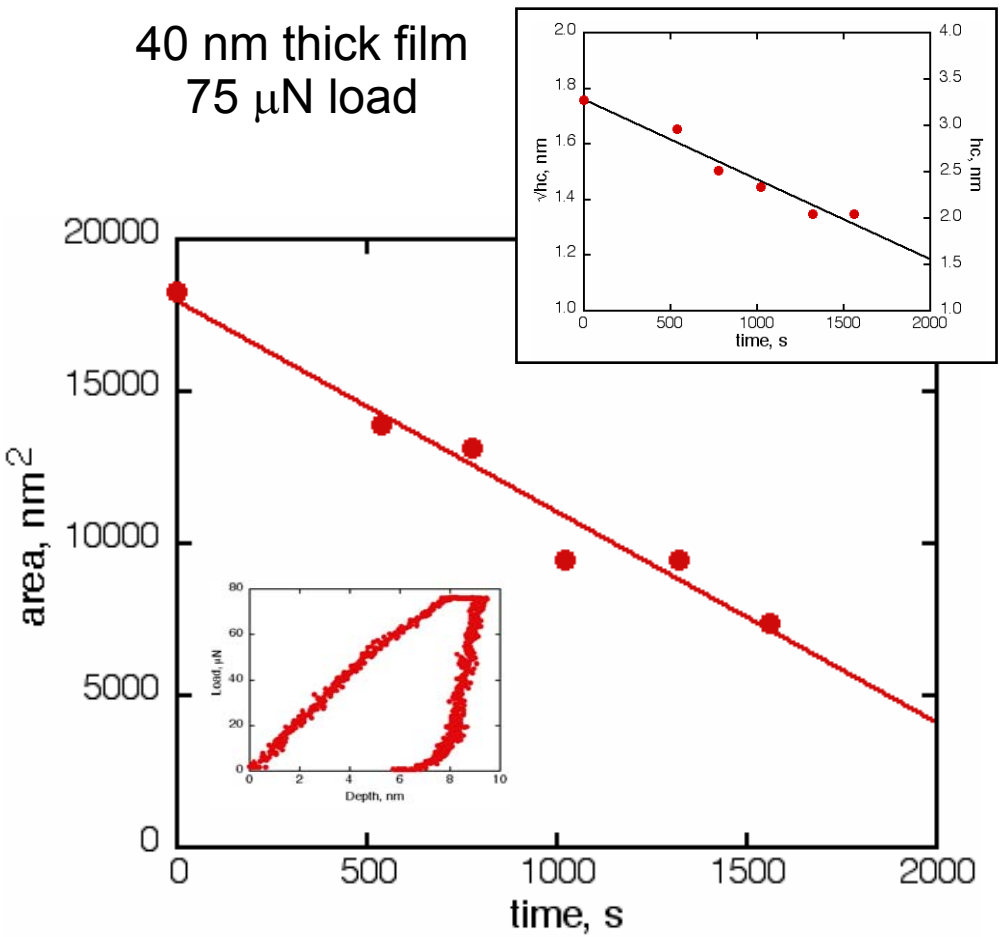


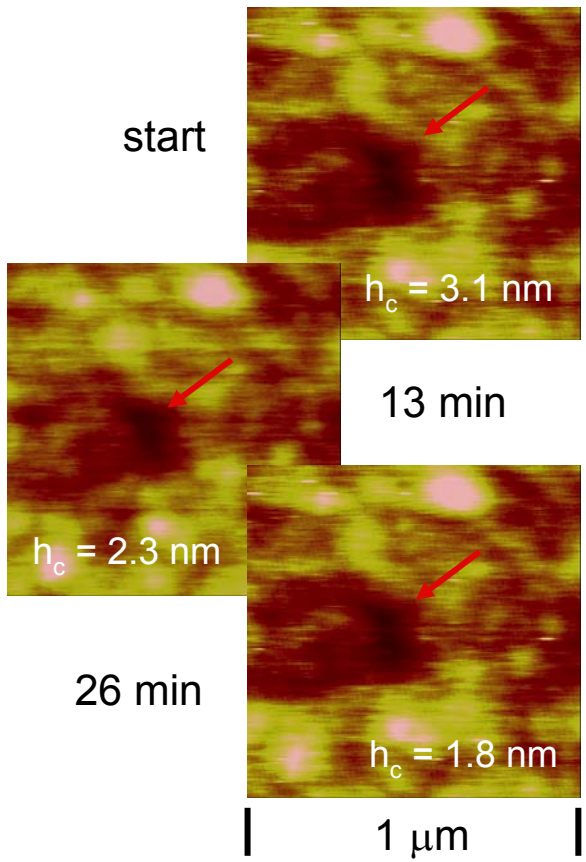
(Zelinski, Huang, Gerberich, 1993)

Imaging the indent defines time-dependent material response as a function of load and displacement

Shallow indentations exhibit time dependent recovery.

40 nm thick film
75 μN load





(M. J. Cordill)

Indentation induced dislocations do not interact strongly with the film microstructure or existing dislocations.

Fracture of Atomic Layer Deposited Tungsten Films

T.E. Buchheit, B.L. Boyce, S. Prasad
T.M. Mayer, R. A. Wind, S.M. George,
J. M. Jungk, W. W. Gerberich

Which property measurements are critical for design of reliable nanomaterials and devices?

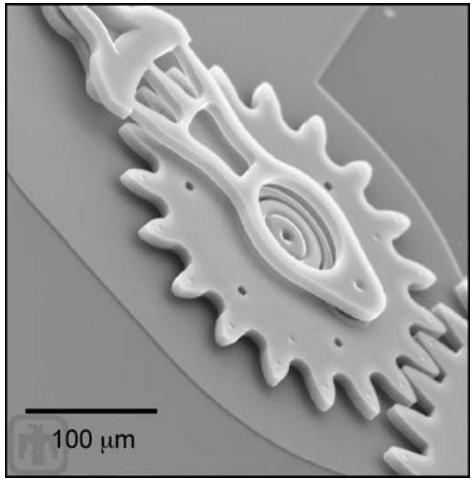
Are existing test methods sufficient?

What is the role of modeling?

What answers does modeling provide?

Materials issues control performance and reliability of microsystem devices.

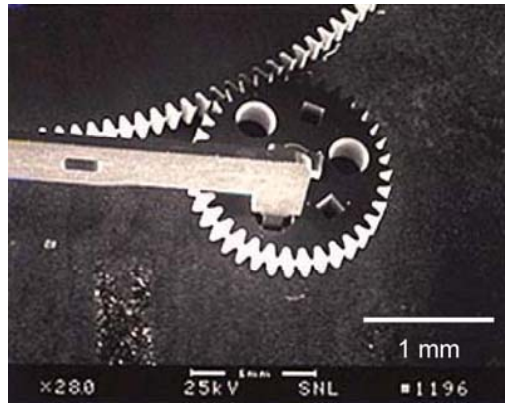
Si Surface Micromachine Technology



- hydrophilic oxides, water adsorption
- adhesion, surface morphology
- chemical stability, ‘dormancy’
- friction/wear
- strength
- high aspect ratios

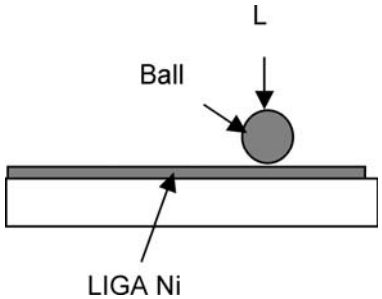
LIGA technology

- electroplated Ni alloys
- surface morphology
- composition, stability
- corrosion
- friction/wear
- strength
- high aspect ratios

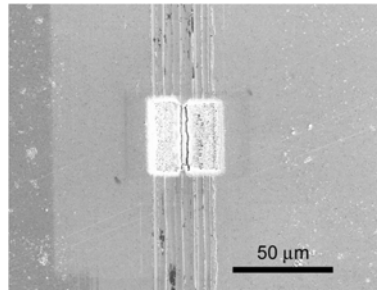


Hard coatings can enhance performance and reliability

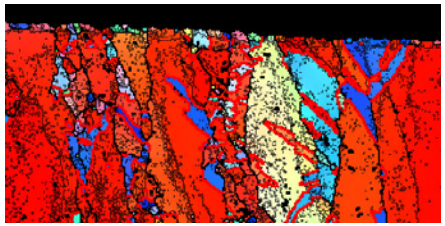
Contact Spring



Wear Scar

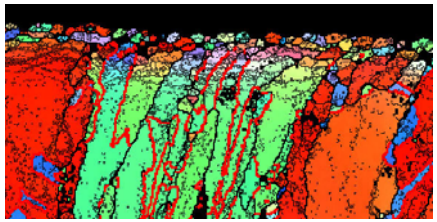


LIGA Ni

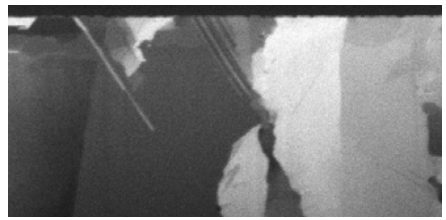


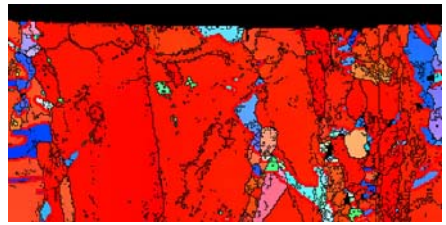
Unworn Surface

LIGA Ni



DLC on LIGA Ni

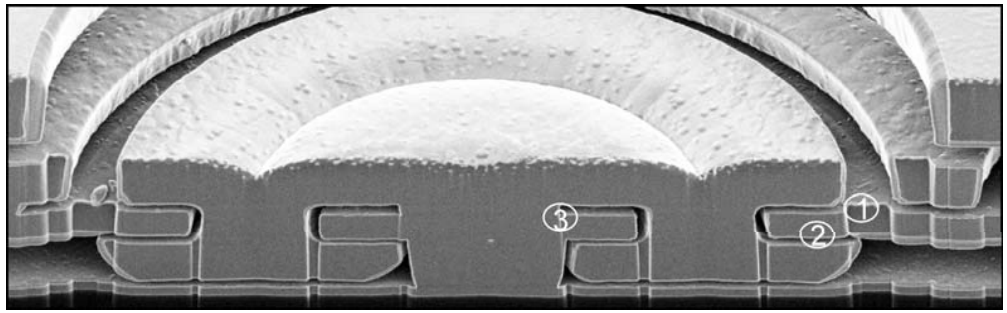




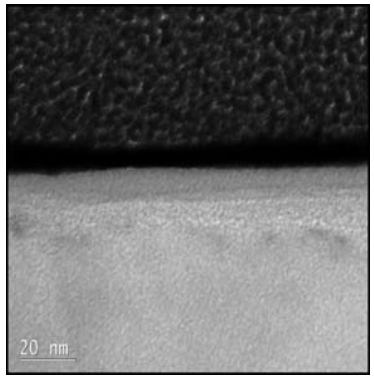
10 gm 1000 cycles

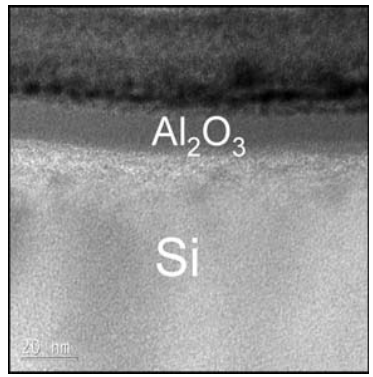
(Prasad, Michael, Christenson, 2003)
(Prasad, et. Al, unpublished results, 2004)

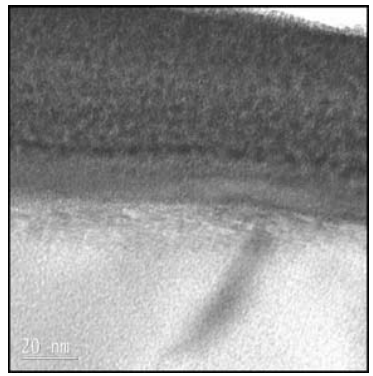
ALD conformally coats very high aspect ratio structures



1. Gear top exposed
2. Gear bottom shadowed
3. Hub interior aspect ratio ~100





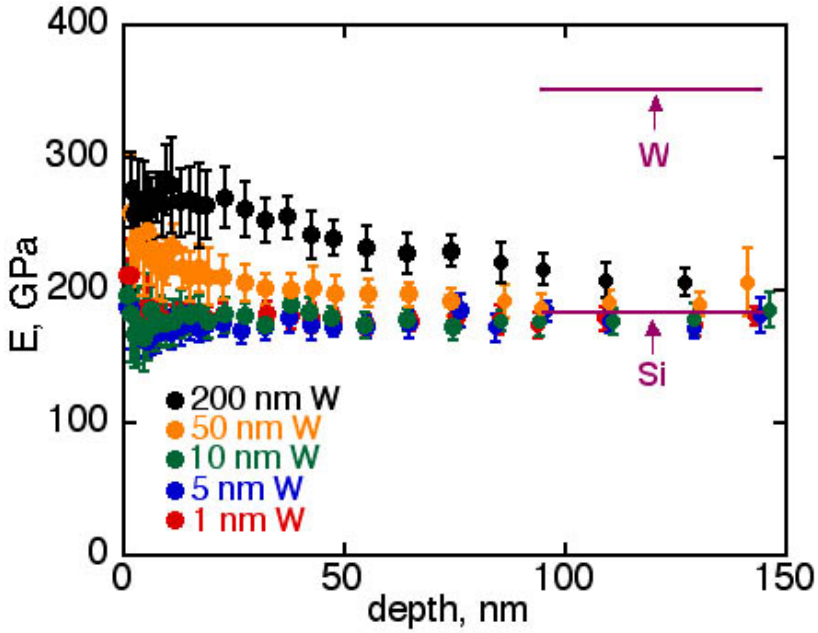


These films are uniform in thickness and amorphous in structure

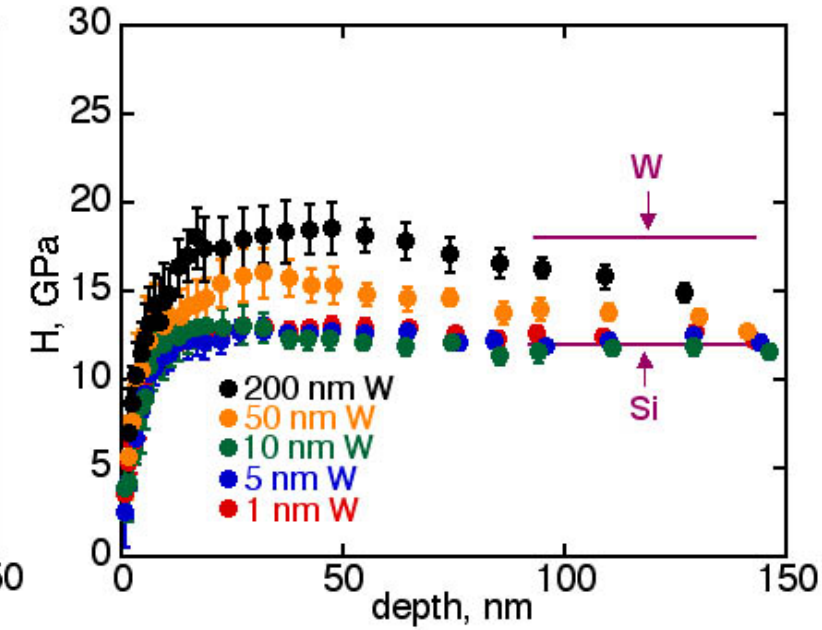
Tungsten films show a strong apparent variation in properties with film thickness.

Tungsten

Modulus



Hardness

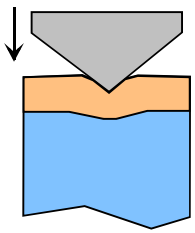


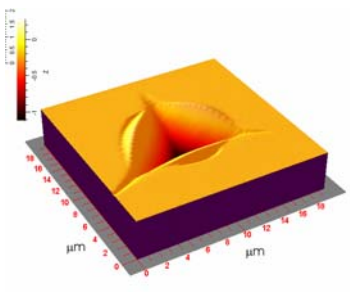
- properties are lower than bulk and sputtered tungsten
- properties of very thin films are masked by substrate effects

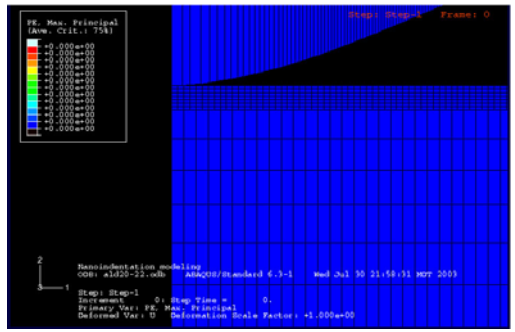
Cannot effectively measure near surface hardness of hard materials

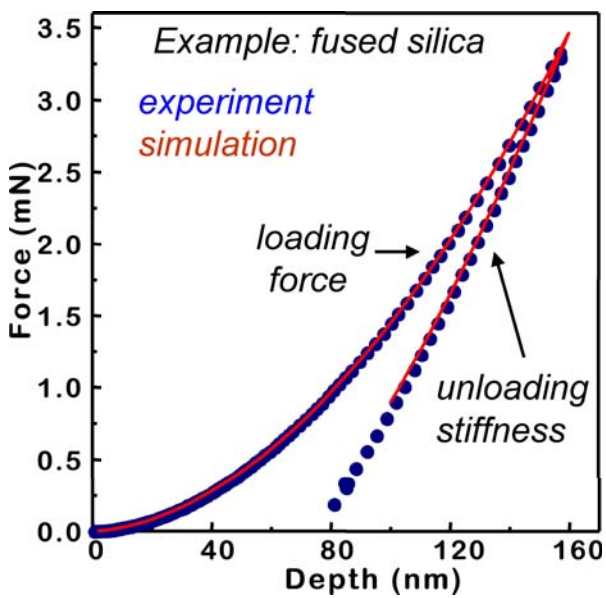
Require elastic modulus, stress and strain data for assessing performance of hard films and structures

EXPERIMENT

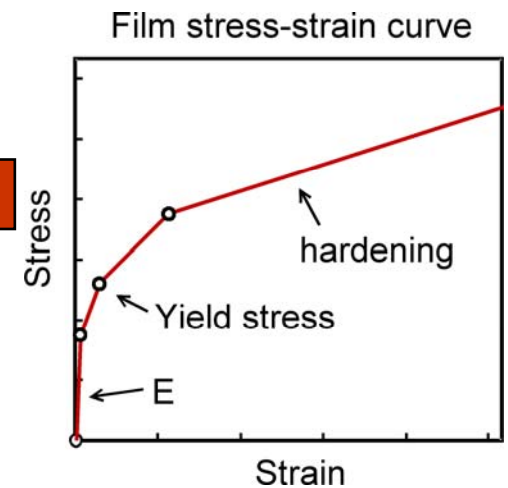








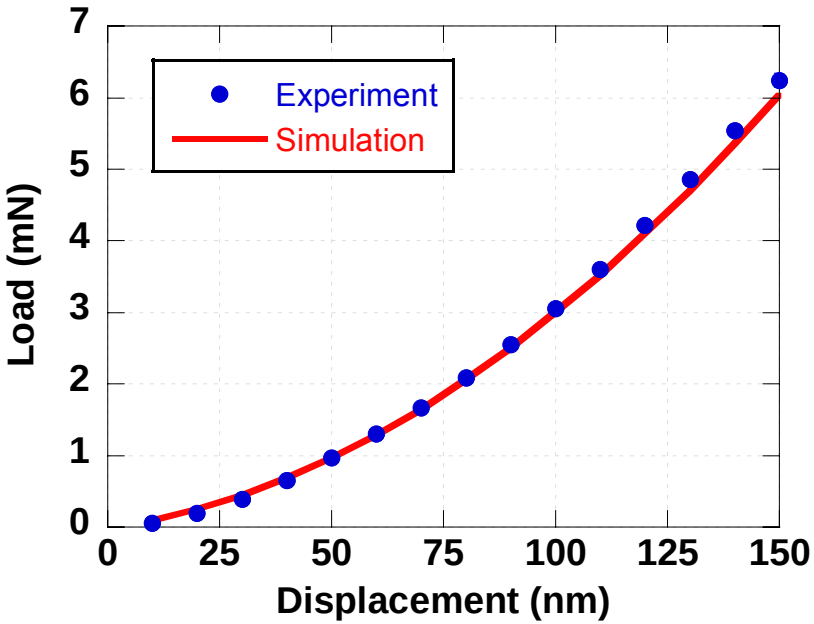
SIMULATION

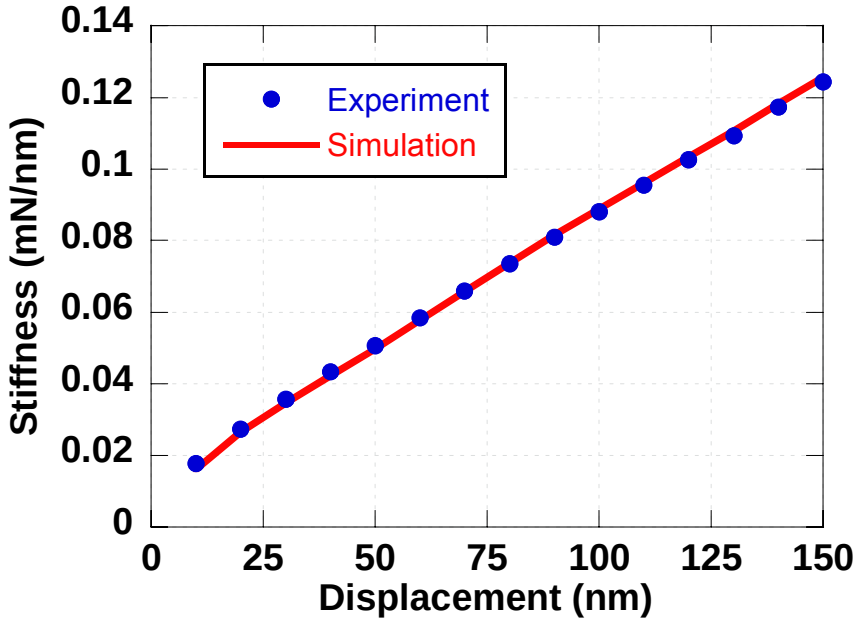


(Jungk, Gerberich, Hoehn, Buccheit, Mayer, Moody, Knapp, 2004)

Finite Element Analysis provides a means to deconvolute film and substrate contributions to measured properties

Finite Element Analysis shows good agreement with near surface measured values for 50 nm thick ALD tungsten

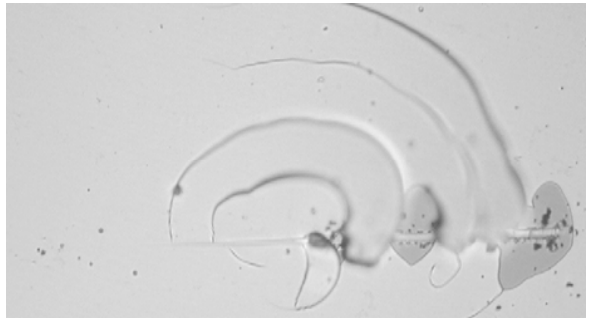




- 50 nm W/Si
- Obtained best fit from:

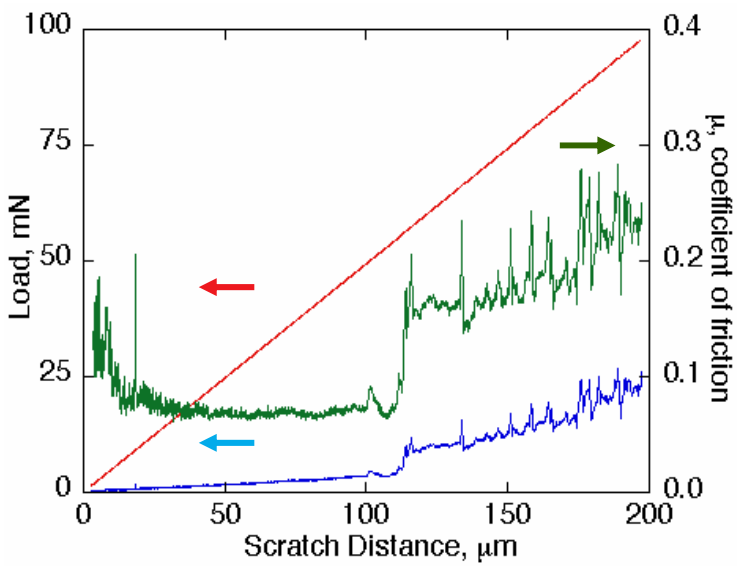
$$E = 204.9 \pm 46.5 \text{ GPa}$$
$$H = 23.6 \pm 8.6 \text{ GPa}$$

Nanoscratch tests are used to assess scratch adhesion



200 nm tungsten ALD Film

1 μm radius conical diamond tip
Applied increasing normal load



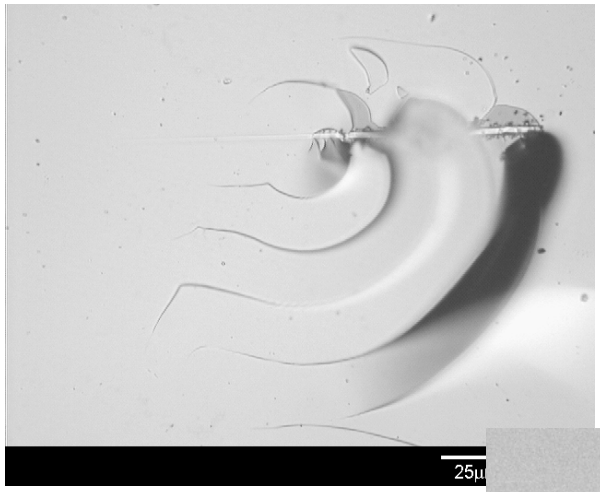
Cracking and delamination clearly evident

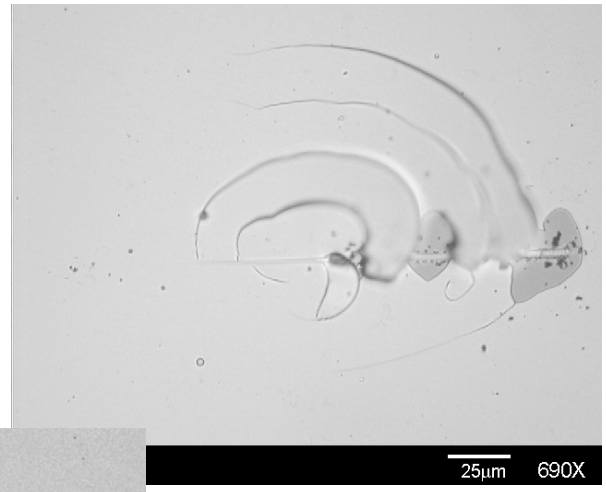
Direct correlation between load and fracture events

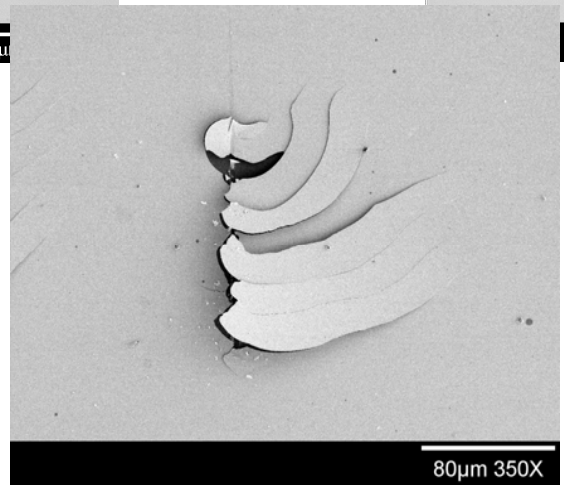
Loads and displacements mirror fracture events

Nanoscratch tests triggered extensive channel cracking and delamination over the surface of the film

200 nm ALD tungsten on polished silicon



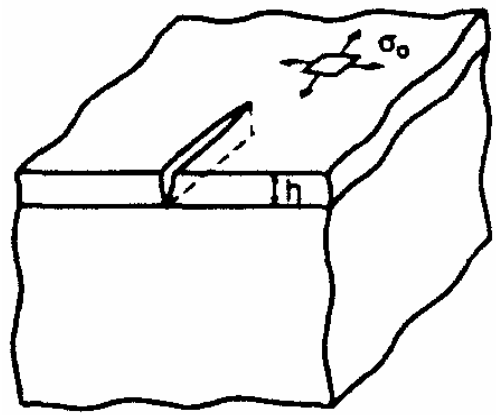




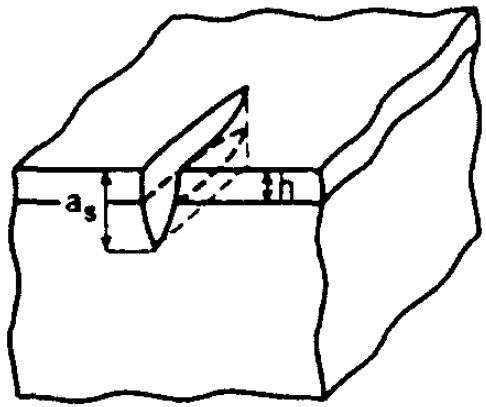
Cracking appears to follow a debond profile

Probe-crack tip interactions, complex stress states, and large probe to film thickness ratios limit direct analysis.

Film Cracking

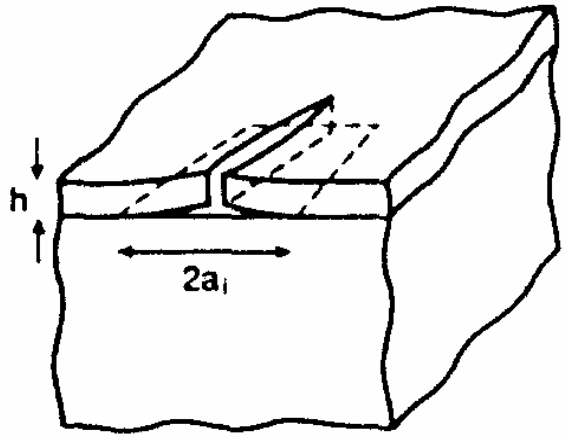


Substrate Cracking



$\Gamma_s < \Gamma_f$

Debonding



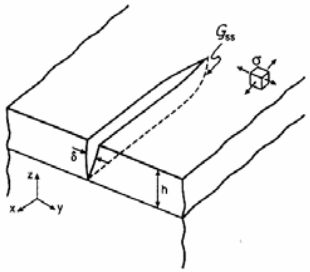
$\Gamma_i < \Gamma_f$

Ye, Suo, Evans (1992)
Hutchinson, Suo (1992)

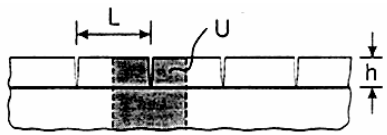
Analytical models can be used to define the conditions for cracking and debonding of films during scratch tests

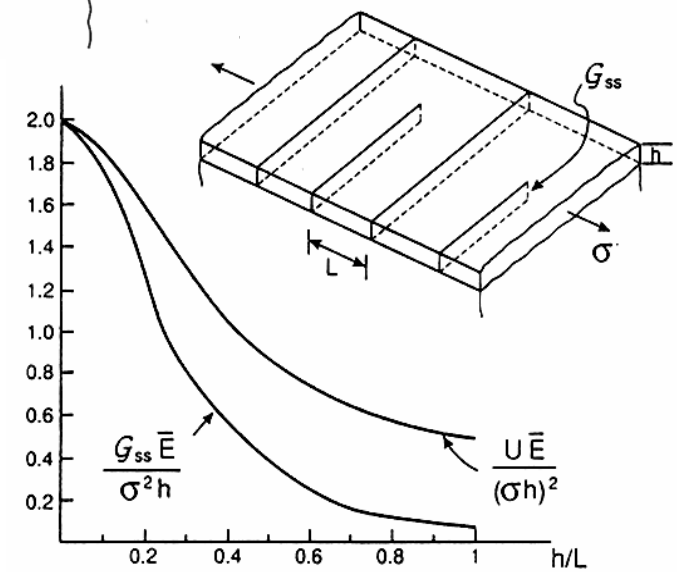
Cracking is driven by high residual tensile stresses

Single Crack



Multiple Cracks





Hutchinson and Suo (1992)
Hu, Thouless, Evans (1988); Thouless (1990)
Ye, Suo, Evans (1992); Beuth (1992)

Deposition at 300˚C leads to a high residual tensile stress,

$$\sigma = 400 \text{ MPa}$$

For a single channel crack, the steady-state fracture energy is,

$$G_{ss} = \sigma^2 h(1-\nu^2)\pi g(\alpha,\beta)/2E$$

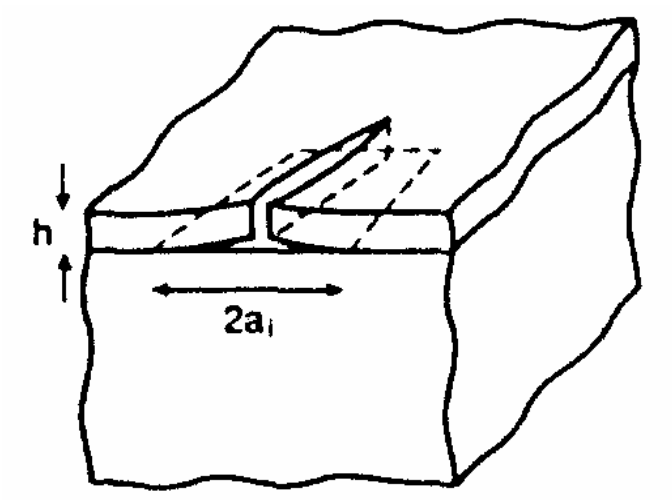
α=0.6, β=0.07
h=200 nm, L=19 μm
g(α,β)=2.3117 (Beuth, 1992)

$$G_{ss} = 0.4 \text{ J/m}^2$$

$$K_I = 0.33 \text{ MPa-m}^{1/2}$$

The single crack solution holds for widely spaced multiple cracks

Interfacial fracture energies are obtained from elastic solutions for edge loaded films



α=0.6, β=0.07, s=0.654
η_i=L/h=50, L=2a
λ_3=0.796, λ_4=0.694

For the interface crack,

$$G_i = \omega_i \sigma^2 h(1-\nu^2)/E$$

where ω_i is given by,

$$\omega_i = \frac{1}{2}\left(\frac{\eta_i}{1+\eta_i}\right)^{1-2s}\left[1+\lambda_3 \exp(-\lambda_4\sqrt{\eta_i}\right]$$

G_i = 50 mJ/m^2

$\psi(\alpha,\beta)$ = 60°

G_I = 30 mJ/m^2

Ye, Suo, Evans (1992)
Hutchinson, Suo (1992)

The interfacial fracture energy shows a weakly bonded film

Widely assumed that debonding follows channel cracking

Why do channel cracks follow an initial circular debond profile?

Why are channel cracks parallel yet so widely spaced?

What is the sequence of fracture?

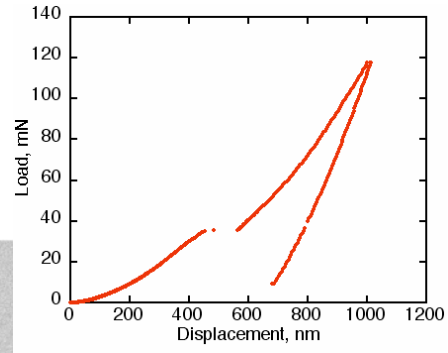
Do analytical elastic mechanics models accurately describe fracture processes?

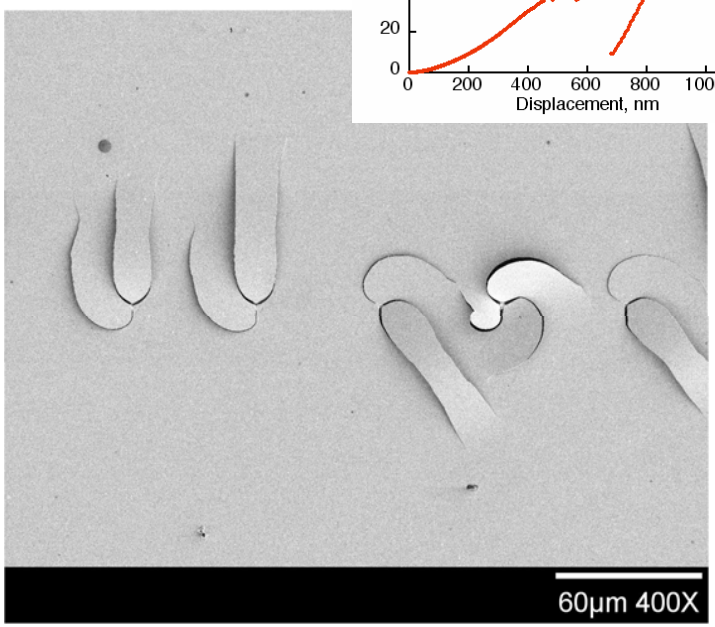
Do model based measured energies reflect actual events?

Acoustic emission provides a potential method for studying individual fracture events

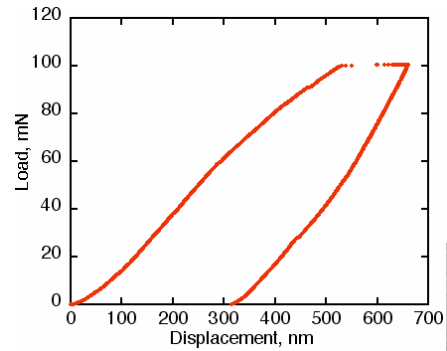
Indentation triggered channel cracking and delamination

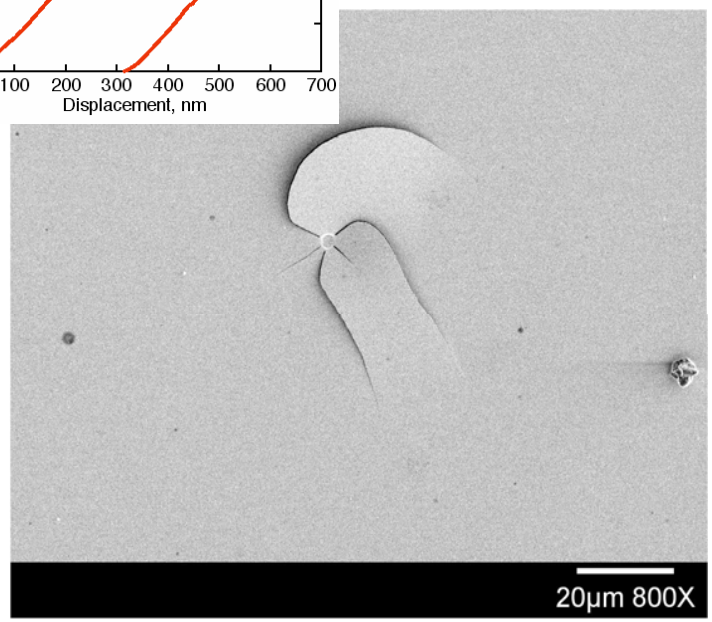
Berkovich





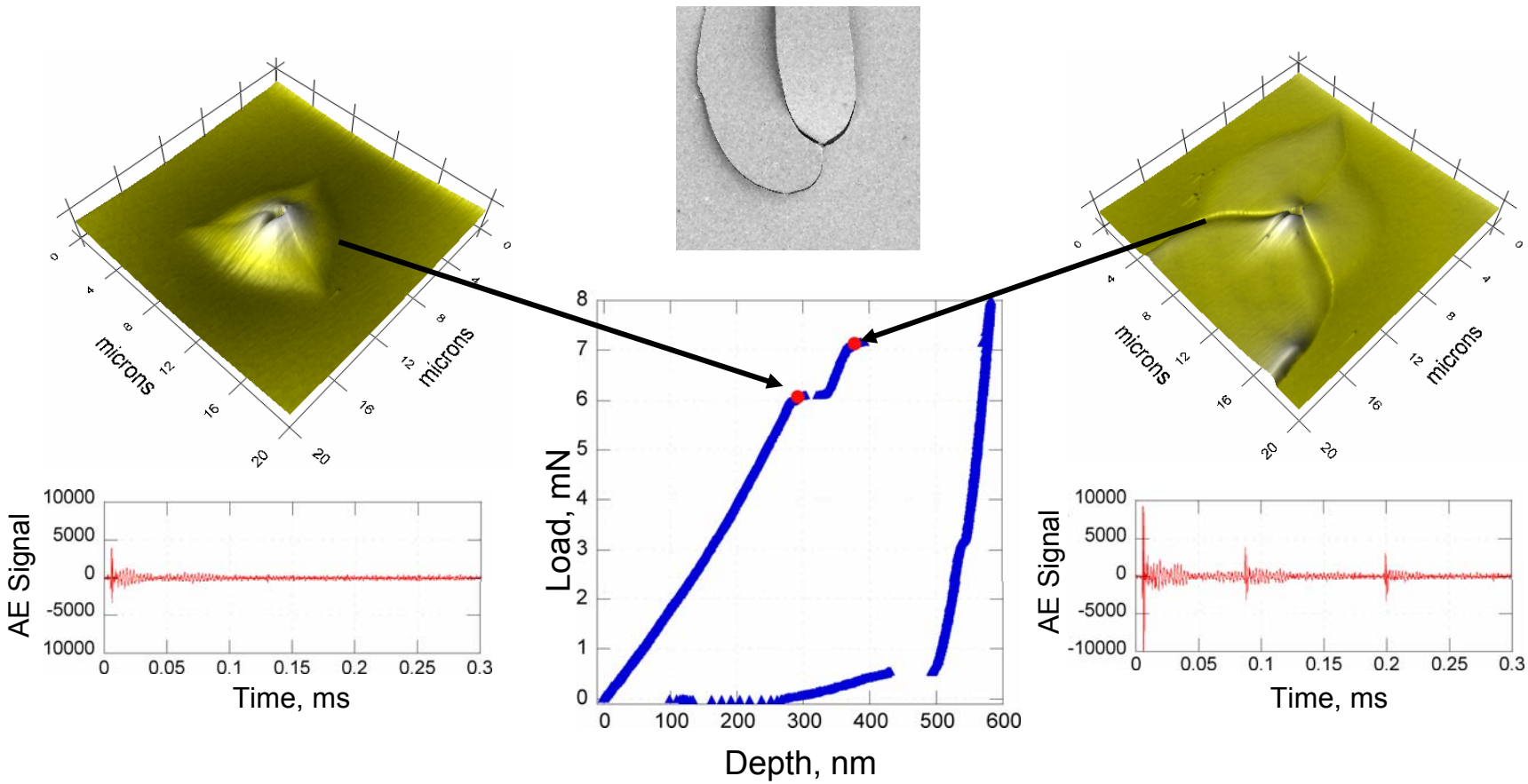
Conical





Fracture occurred with through thickness indentation independent of load.

Acoustic emission events occur with film delamination and fracture.



First AE event occurs with film delamination.

Subsequent events accompany channel cracking

Fracture begins by film delamination followed by channel cracking

Summary

Deformation at very small scales controls adhesion and fracture of thin ductile films

Substrate effects can mask the properties of thin hard films

Probe-crack tip interactions, complex stress states, and large probe to film thickness ratios limit quantitative analysis of nanoscratch tests

Analytical modeling and elastic concepts can define conditions for fracture in select cases

Experimental and theoretical approaches must be combined for quantitatively correct properties and physically accurate processes

Challenges in assessing the role of adhesion and fracture on reliability of nanostructured materials and devices

Direct imaging of processes during tests is the ideal

Sample size and process length scales often do not permit intrusive techniques in nanostructured materials or devices.

Limitations demand clever and innovative test techniques

Need advanced material modeling of crack tip processes

- creative analytical models
- discretized dislocation models
- atomisitc modeling under realistic conditions

Evolutionary advances will be needed for measuring adhesion of structures at nano size scales.

Acknowledgments

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