Dislocation Nucleation and Multiplication in Small Volumes: The Onset of Plasticity during Indentation Testing

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While classical studies of dislocation behavior have focused on the motion and multiplication of dislocations, recent advances in experimental methods allow studies that probe relatively dislocation-free volumes of materials. When the dislocation concept was initially developed 75 years ago, researchers were not easily able to examine the ultra-fine size scales of materials that would allow the onset of plasticity to be examined. This paper will review some recent developments in the area of incipient plasticity in materials and provide a historical context for current interest in testing mechanical behavior at small scales.

INTRODUCTION

In 1934, three separate papers by G.I. Taylor, E. Orowan, and M. Polyani were published that proposed mechanisms for plasticity in crystalline solids that was controlled by what we now refer to as an edge dislocation.[1–3] Though other concepts for slip in crystals had previously been proposed, it is these papers that effectively form the basis from which dislocation theories and models have been developed. Five years later, the screw dislocation was proposed by J.M. Burgers.[4] Direct observation of dislocations in the electron microscope did not occur until two decades later.[5] One of the motivations for the initial work was the fundamental question of how metals permanently deformed at applied stresses that were orders of magnitude lower than those estimated to be required to break bonds along planes of atoms, the so-called "theoretical shear stress." Many models for the shear stress needed to simultaneously displace a plane of atoms have been proposed, but in general the classical theoretical shear strength of a material is often approximated by $\mu/30$ to $\mu/100$,[6] where μ is the shear modulus.

The motion of dislocations allows for permanent deformation at much lower stresses than the theoretical shear stress. Researchers trying to achieve high-strength materials have traditionally focused on increasing the resistance to dislocation motion, but in the 1950s the work of C. Herring and J.K. Galt[7] suggested another route. If dislocation-free metals could be made, they should have extremely high strengths, as the nucleation of dislocations in a perfect crystal would require application of stresses equal to the theoretical shear stress. S.S. Brenner summarized the results of several different materials that had been fabricated as whiskers, thin metallic crystals on the order of 1 µm in diameter, and showed definitively that their strength approached the theoretical shear strength of materials.[8]

How would you…

…describe the overall significance of this paper?

This paper summarizes some of the state-of-the-art issues in using nanoscale mechanical testing to assess dislocation nucleation. Researchers in this area are using new techniques to address issues that arose as early as 75 years ago with the initial dislocation theories.

…describe this work to a materials science and engineering professional with no experience in your technical specialty?

Many of our estimates of theoretical shear stress for dislocation nucleation and multiplication can now be directly measured using new techniques. However, care must be taken to ensure the assumptions made during nanoindentation are appropriate for the given experiment.

…describe this work to a layperson?

For 75 years we have not been able to directly measure some of the properties of the defects that are responsible for materials that deform permanently. With the techniques described in this paper, developed over the past 10–20 years, we are now able to begin to accurately measure these fundamental properties of many materials, particularly metals.

INDENTATION TESTING

Classical indentation testing, including but not limited to Rockwell, Vickers, Knoop, or Brinell tests, has been used widely in the assessment of mechanical properties of materials. One critical issue in these macroscopic investigations of the strength of materials is the size of the sampled volume. Indentation testing usually probes a sample over length scales of millimeters (Brinell) to tens of micrometers (Vickers). While indentation testing samples a much smaller volume than traditional tensile or compression tests, the sampled volume is such that, in most metals, the probe will have interacted with a significant number of pre-existing dislocations. Therefore, hardness values are often related to the flow stress in solids because of the uniformity of the deformation around the indentation;[9,10] the classic models suggest, for instance, that the mean pressure exerted during a Vickers hardness test is about three times the compressive stress required to generate 8% strain in the sample.[11] Hardness testing is very important for nondestructive assessment of the mechanical properties of materials, yet the sampled volume is still large enough that measurements of dislocation-free solids, which could be used to monitor the onset of plasticity, are unlikely.

In the 1960s, N. Gane and F.P. Bowden carried out indentation testing in an electron microscope on electropolished surfaces of gold, copper, and aluminum, and were the first to observe what is now often referred to as an "excursion" or "pop-in."[12] A fine tip was pressed into the gold surface, but no permanent penetration was observed until a critical load was reached, at which point the tip suddenly and rapidly penetrated the surface. By selecting a small tip, and indenting a well-annealed sample, the volume probed by the tip was likely on the order of the spacing between dislocations. Using an electrical resistance technique, J.B. Pethica and D. Tabor[13] performed similar indentation experiments in ultra-high vacuum. They observed that a material could withstand large stresses, possibly reaching the theoretical shear stress, when an oxide layer was present on top of the indented surface. These pioneering experiments paved the way for studies of the initiation of plasticity with nanoindentation.

CURRENT TESTING METHODOLOGIES

There are two basic types of experiments that focus on determining the theoretical strength of metals. In the first case the sample size must be small enough to be dislocation free, but the tests can be tied to uniform stress states (analogous to the tensile testing of whiskers. In the second case (indentation testing), bulk materials with low dislocation densities may be tested, which allows more flexibility in material selection. The drawback to indentation is that the stress state is more complicated than in a uniaxial tension, compression, or torsion test. However, as commercially available nanoindentation instruments have become more affordable and user-friendly, nanoindentation experiments are the most common way to identify the stresses at which plasticity begins.

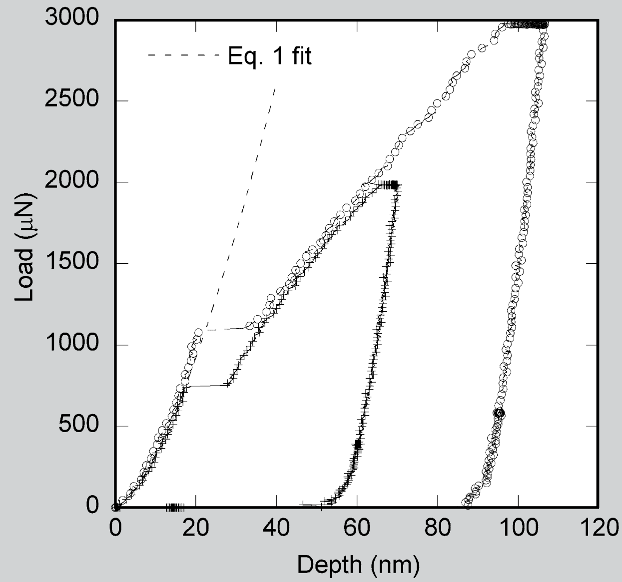

Figure 1. Typical experimental results for two indentations into a {100} tungsten single crystal, showing initial elastic loading. The variation in "pop-in" load and Hertzian fit (Equation 1) can be seen. In this case the inferred tip radius is approximately 600 nm; the probe used is a blunted Berkovich-type diamond. Note the similarity in the loading curves once the yield point has been exceeded.

In a load-controlled nanoindentation experiment, the onset of plasticity is seen as a sudden displacement (pop-in), as shown in Figure 1. This type of behavior has often been linked to dislocation nucleation[14–18] and dislocation source activation.[19,20] One of the major challenges in assessing this elastic–plastic transition during nanoindentation is the difficulty in ascribing the behavior to any one mechanism; some of the proposed mechanisms include: homogeneous dislocation nucleation,[21] the activation of well-spaced dislocation sources,[22] the activation of a point defect source (e.g., a vacancy),[20] the fracture of a surface film,[23–26] or interaction of a surface film with the underlying dislocation structure.[27]

The ability to test small volumes of materials has led to a common observation: "smaller is stronger." Often this adage has been linked to indentation tests, such as the case of the "indentation size effect."[28] Recently, researchers have been able to regularly and reproducibly fabricate micrometer-scale specimens from more macroscopic volumes of materials using focused ion beam (FIB) machining. While most compression testing of these microscale structures has been carried out using nanoindentation platforms,[29] there are also examples of FIB-machined tensile specimens with FIB-machined tensile grips.[30] Focused ion beam machining has also been used to fabricate structures such as pillars of diameter from 200 nm to 40 μm for compression testing.[31]

Using a combination of thin film deposition and micromachining, it is possible to carry out in-situ observations of deformation in the transmission electron microscope (TEM) during indentation.[32] For example, by adding an instrumented indentation system to the TEM, correlations were made between the nanoindentation load–depth relationships and specific dislocation activity behavior.[33] This type of tool is not limited to testing unstructured thin films, but can also be used to examine pillars in compression and even more complicated structures such as nanoporous gold, where dislocations have been observed in ligaments on the order of 5 nm in diameter.[34]

These experiments have greatly increased our understanding of dislocation mechanisms in sub-μm^3 volumes under high applied stresses. While there is significant appeal in using the in-situ and the FIB-machined structures for subsequent interpretation of the stresses required for dislocation nucleation, the prevalence of instrumented indentation equipment suggests that there is still room to explore incipient plasticity mechanisms using nanoindentation of relatively dislocation-free volumes of materials. Therefore, the remainder of this paper will focus on one common method: using a nominally spherical probe to indent a nominally uniform surface of a material. This method is not limited to testing metals; the technique is applicable to ceramics[35,36] and even organic crystals such as sucrose.[37]

INTERPRETATION OF NANOINDENTATION EXPERIMENTS

Nanoindentation instruments measure the forces and displacements remotely imposed on a one-degree-of-freedom (1-DOF) mechanical probe. The probe is suspended by springs designed to approximate the 1-DOF system as closely as possible. Typically, displacements are measured by gap-closing capacitance measurements; the theoretical resolution of these measurements can be sub-picometer but are practically limited, by electronic noise and coupling to ambient vibration, to about 0.5 nm. Forces are typically imposed through electromagnetic or electrostatic means; the accuracy in the measured mutual force between probe and sample is thus dependent not only on the resolution of the force-application method, but the displacement noise, accurate stiffness measurement of the suspension, and the ability to detect the point of first contact (the "zero point"). Typical force noise floors are 1 μN–10 μN. These measurement capabilities enable the detection of elastic-plastic transitions with probes of very small radii (approximately 100 nm).

The initial loading of a nanoindentation is greatly dependent on the surface preparation. Indentations into mechanically polished samples (with consequent high dislocation densities) often do not exhibit a distinct transition from elastic to plastic deformation. However, when annealed and electropolished surfaces are examined, this transition becomes much more common. For the remainder of this paper the focus will be on samples with low dislocation densities. It has been noted that the load–depth curve, the "mechanical fingerprint,"[38] follows the same basic shape for a given indenter once there is a well developed plastic zone. This means that after the excursion in depth occurs (for a load controlled instrument), the load–depth curves often follow each other for a given material. An exception to this behavior is the staircase yielding phenomena,[39,40] where multiple bursts occur until the "fingerprint" curve is reached after several steps.

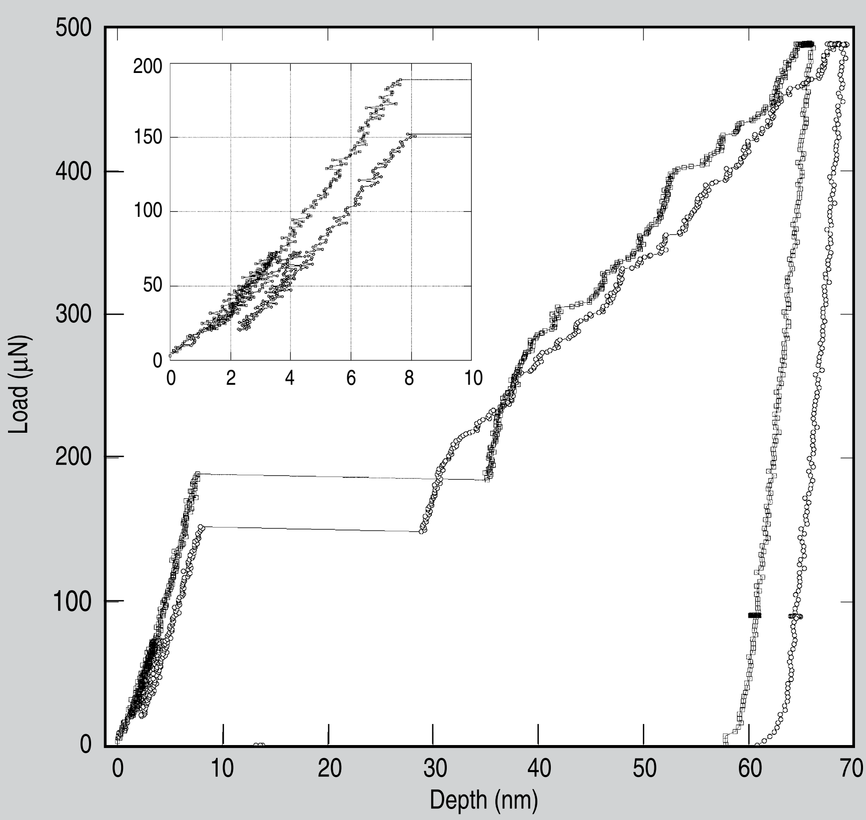

Figure 2. Two indentations into a {111} nickel surface with a partial unloading sequence carried out at the start of the loading. These indentations were made at a constant loading rate to 70 μN, the load was reduced at a constant rate to 25 μN, and the load was increased again at the same rate to the maximum load of 500 μN. The inset shows the difference between a purely elastic indentation and one which exhibits minor, but measureable, plastic deformation prior to the "pop-in" event. While it is possible to fit the elastic-plastic load-depth curve to the elastic Hertzian model, it will produce an inaccurate tip radius.

In typical indentation tests designed to examine the initial yield events, the elastic properties of the material under test are known beforehand. The initial contact is also believed to be purely elastic. The elastic contact condition can be verified by carrying out partial unloading experiments, as shown in Figure 2. Care must be taken to ensure the behavior is elastic, as it is possible to observe pop-in excursions during loading even when the loading is not elastic, as shown in the inset of Figure 2. If the behavior is indeed elastic, then the Hertzian approximation for elastic contact between two spheres may be used. The Hertzian load–depth relationship is

$$P = \frac{4}{3}\bar{E}_R R^{1/2} h^{3/2} \qquad (1)$$

where P is the load, h is the depth, and $\bar{E}_R$ is the reduced plane-strain modulus between indenter (I) and specimen (S), $\bar{E}_R = \left[\left(1-\nu_I^2\right)/E_I + \left(1-\nu_S^2\right)/E_S\right]^{-1}$. The effective radius, $R = \left[1/R_I + 1/R_S\right]^{-1}$, reduces to the radius of the indenting probe if the specimen surface is flat. If the radius is known, analytical expressions[11] may be used to find field values of stresses in the body. The maximum shear stress at a Hertzian contact, τ, is directly beneath the surface with a value of

$$\tau = 0.31\left[\frac{6\bar{E}_R^{\,2}}{\pi^3 R^2}\right]^{1/3} P^{1/3} \qquad (2)$$

If there is not a distinct yield point, gradual yielding occurs in which the load–depth curve begins to deviate from the predicted elastic loading curve at a load of[11]

$$P_y = \frac{\pi^3 R^2}{6\bar{E}_R^{\,2}}\left(1.6\sigma_y\right)^3 \qquad (3)$$

where σ_y is the yield stress in simple compression (or tension). Gradual yielding at an elastic-plastic contact has been demonstrated experimentally with large (70 μm radius) spherical indenters.[41]

THE SPHERICAL PROBE APPROXIMATION

In the previous section, it was assumed that the probe radius of curvature could be inferred from an elastic loading condition. Many of the plastic yield initiation studies noted above were studied with conical probes that are intentionally manufactured to have a spherical cap at the apex. These "conospherical" probes can be fabricated with radii as small as 1 μm. Although this is very small, access to the nanometer length scales needed to find a dislocation-free volume may require even smaller radii. Experimentalists often take advantage of the imperfect manufacture of nominally sharp geometries, such as the three-sided pyramidal Berkovich probe. A common geometric view is that the imperfection at the apex is a spherical cap that smoothly merges with the pyramidal planes. This idea forms the basis of many analytic area functions for nanoindentation hardness and modulus measurement.[42,43]

There are two frequent assumptions in yield-point experiments: that the apexes of indenting probes are reasonably spherical; and then, that Hertzian analysis (Equations 1 and 2) is appropriate to analyze the experiment. If so, a fit of Equation 1 to load–depth data up to the pop-in event may be used to estimate the effective radius of the probe. While most manufacturers of sharp probes claim the ability to produce an apical radius less than 50 nm, this is only a rough estimate. Blunting of the probe occurs with each new indentation experiment, especially during the first experiments when the probe is sharpest. Measurement of the probe radius by microscopic means is difficult and time-consuming (especially when the radius is very small), which is why inferential measurement of the probe radius by curve-fitting to the Hertzian load–depth relationship is a very commonly employed technique.

There are indications that Hertzian analysis may not always be justified. For example, finite-element analysis (FEA) has shown that an irregular probe will produce an elastic load–depth relationship that could certainly be perceived as originating from a spherical contact,[44] although the real probe geometry was not spherical at all. That is to say, although the load–depth data could reasonably be described by a fit in the manner of Equation 1, the contacting surfaces did not fit the Hertzian criteria (specifically, that of contacting paraboloids). An FEA simulation showed that the irregular probe generated shear stresses in the body that were significantly different, both in magnitude and location, from those generated with a true spherical probe. Elastic contact by an irregular probe focuses the indentation force into a smaller volume, and thereby increase the maximum shear stress in the body over that produced by a spherical contact.

In an attempt to experimentally address the effects of indenter probe irregularity, we have measured the near-apex shape of a commercially purchased Berkovich probe using scanning probe microscopy (SPM) in a recent study.[45] The radius of curvature at the tip was estimated by analysis of the SPM data to be 121 nm ± 13 nm (mean ± standard deviation of 8 measurements). This probe, shown in Figure 3, was then used to indent both tungsten and nickel single crystals (the data from the nickel is plotted in Figure 2; the indentations shown in Figure 1 were performed with another probe). After six purely elastic nanoindentations on a {100} face of a tungsten crystal were performed, the tip radius was estimated as 163 nm ± 13 nm, a value ≈ 35% larger than the SPM-derived measurement.

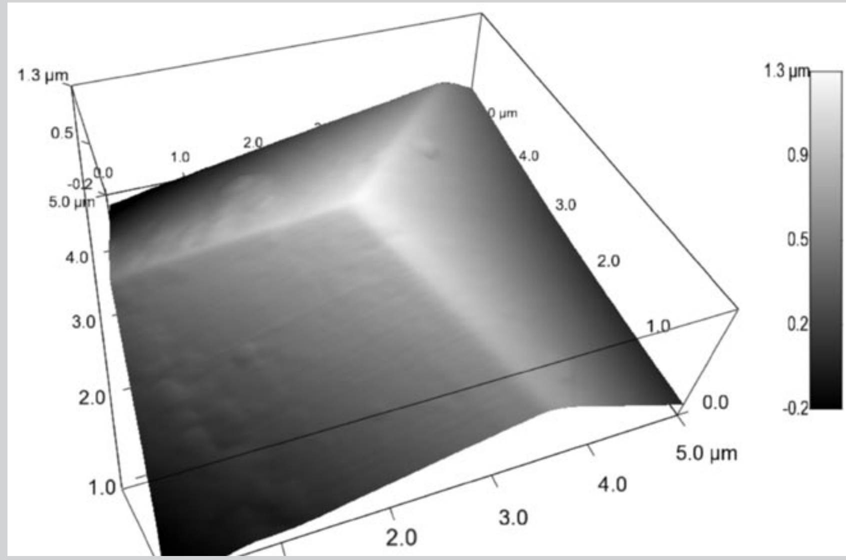

Figure 3. A scanning probe microscopy image of the probe used to carry out the indentations shown in Figure 2. This three-sided Berkovich geometry shows the tip rounding that is unavoidable during manufacture.

Fully three-dimensional, FEA simulation of indentation by the SPM-measured probe was able to recreate the experimental load–depth curves. Furthermore, we found a surprising result from the simulation that neatly paralleled the experiment: when a sphere of known radius is used in the virtual indentation, the radius perceived by Hertzian analysis was about 40% larger! That is to say, the contact is stiffer than the Hertzian relationship would predict; and by curve-fitting to find the radius as if it were unknown, we would have guessed that the sphere is larger than it really was. What has happened is that the limits of linear elastic analysis have been surpassed at the point of incipient yield. Perhaps we should have not been so surprised; Brenner's 1956 iron-whisker experiments also demonstrated non-linear elasticity just before the onset of plastic yield. The total effect is that the Hertzian analysis likely underestimates the shear stresses that drive initial yielding, due to an unknown combination of probe irregularity and non-linear elasticity that causes overestimation of the radius of curvature of the probe.

WHAT IMPACTS THE YIELD POINT?

A significant number of experiments have been published using the yield point phenomena. These include studies at various temperatures,[46,47] metallic impurities,[48] time–stress combinations to examine incipient plasticity,[18] body-centered cubic metals,[16,49] face-centered cubic metals,[39] differing crystal orientation,[50] existing dislocation densities,[22,33] and hydrogen effects in metallic alloys[51,52] to name a few of the many possible parameters. This list neglects studies that have focused on the fracture of thin surface oxide films. Indentation yield-point experiments will always suffer from the fact that they destroy evidence of pre-existing defects, and so the determination of the underlying cause of the plasticity burst must be found inferentially. However, a consensus is emerging that the underlying material is the critical factor in causing the yield point to occur. Dislocation nucleation (be it a homogeneous or heterogeneous event which triggers the avalanche of dislocations) in a solid can, indeed, be experimentally assessed in a wide range of materials.

Another attractive feature of the nanoindentation technique in the study of the onset of plasticity is the convergence in length scales between experiments and atomistic simulations. For instance, the ability to computationally assess the effects of surface steps[53] and grain boundaries[54] with dislocations and stress fields can provide insight into strengthening mechanisms in solids. While computational simulations are currently limited to short time scales when compared to experimental studies, there is clearly hope for continued interaction in this area toward understanding the stresses required to initiate plasticity.

CONCLUSIONS

Nanoindentation testing to probe fundamental dislocation behavior has made significant advances in the past decade. Rather than studying the motion of existing dislocations, yield point phenomena during nanoindentation provides an opportunity to study the creation of dislocations, along with all the possible associated interactions between structure and defects. The conventional Hertzian assumptions provide conservative estimates of the shear stresses needed to generate plasticity in low dislocation density solids. Further studies will hopefully continue on the path, laid out 75 years ago, toward understanding the mechanisms responsible for generating dislocations in solids.

References

1. G.I. Taylor, *Proc. Roy. Soc. A*, 145 (1934), pp. 362–387.
2. E. Orowan, *Zeit. Phys.*, 89 (1934), pp. 605–613, 614–633, 634–659.
3. M. Polanyi, *Zeit. Phys.*, 89 (1934), pp. 660–664.
4. J.M. Burgers, *Proc. Kon. Ned. Akad. Wetenschap.*, 42 (1939), pp. 293, 378.
5. P.B. Hirsch, R.W. Horne, and M.J. Whelan, *Phil. Mag.*, 1 (1956), p. 677.
6. A.H. Cottrell, *Dislocations and Plastic Flow in Crystals* (Oxford, U.K.: Oxford Press, 1953), pp. 11–12, 53–54.
7. C. Herring and J.K. Galt, *Phys. Rev.*, 85 (1952), pp. 1060–1061.
8. S.S. Brenner, *Science*, 128 (1958), pp. 569–575.
9. L.E. Samuels and T.O. Mulhearn, *J. Mech. Phys. Solids*, 5 (1957), pp. 125–134.
10. T.O. Mulhearn, *J. Mech. Phys. Solids*, 7 (1959), pp. 85–96.
11. K.L. Johnson, *Contact Mechanics* (Cambridge, U.K.: Cambridge University Press, 1999).
12. N. Gane and F.P. Bowden, *J. Appl. Phys.,* 39 (1967), pp. 1432–1435.
13. J.B. Pethica and D. Tabor, *Surf. Sci.*, 89 (1979), pp. 182–190.
14. A.B. Mann and J.B. Pethica, *Appl. Phys. Lett.*, 69 (1996), pp. 907–909.
15. W.W. Gerberich et al., *Acta Mater.*, 44 (1996), pp. 3585–3598.
16. S.A. Syed-Asif and J.B. Pethica, *Phil. Mag. A*, 76 (1997), pp. 1105–1118.
17. D.F. Bahr, D.E. Kramer, and W.W. Gerberich, *Acta Mater.,* 46 (1998), pp. 3605–3617.
18. Y. Chiu and A. Ngan, *Acta Mater.,* 50 (2002), pp. 1599–1611.
19. J. Bradby, J. William, and M. Swain, *J. Mater. Res.*, 19 (2003), pp. 380–386.
20. C.A. Schuh, J.K. Mason, and A.C. Lund, *Nature Mater.*, 4 (2005), pp. 617–622.
21. C.L. Kelchner, S.J. Plimpton, and J.C. Hamilton, *Phys. Rev. B,* 58 (1998), pp. 11085–11088.
22. A.A. Zbib and D.F. Bahr, *Metal. Mater. Trans. A,* 38 (2007), pp. 2249–2255.
23. D. Rodriquez–Marek, M. Pang, and D.F. Bahr, *Metal. Mater. Trans. A*, 34A (2003), pp. 1291–1296.
24. M. Pang and D.F. Bahr, *J. Mater. Res.*, 16 (2001), pp. 2634–2643.
25. E. Weppelmann and M.V. Swain, *Thin Solid Films*, 286 (1996), pp. 111–121.
26. S.V. Hainsworth, M.R. McGurk, and T.F. Page, *Surf. Coat. Tech.*, 102 (1998), pp. 97–107.
27. D.E. Kramer, K.B. Yoder, and W.W. Gerberich, *Phil. Mag. A* , 81 (2001), pp. 2033–2058.
28. P.M. Sargent, *J. Mater. Sci. Lett.*, 8 (1989), pp. 1139–1140.
29. C.A. Volkert and E.T. Lilleodden, *Phil. Mag.*, 86 (2006), pp. 5567–5579.
30. D. Kiener et al., *Acta. Mater.*, 56 (2008), pp. 580–592.
31. M.D. Uchic et al., *Science*, 305 (2004), pp. 986–989.
32. A.M. Minor et al., *J. Mater. Res.,* 19 (2004), pp. 176–182.
33. A.M. Minor et al., *Nature Mater.,* 5 (2006), pp. 697–702.
34. Y. Sun et al., *JOM*, 59 (9) (2007), pp. 54–58.
35. S.V. Hainsworth, T. Bartlett, and T.F. Page, *Thin Solid Films*, 236 (1993), pp. 214–218.
36. Y. Gaillard, C. Tromas, and J. Woirgard, *Phil. Mag. Letters*, 83 (2003), pp. 553–561.
37. K.J. Ramos and D.F. Bahr, *J. Mater. Res.,* 22 (2007), pp. 2037–2045.
38. S.V. Hainsworth, H.W. Chandler, and T.F. Page, *J. Mater. Res.,* 11 (1996), pp. 1987–1995.
39. S.G. Corcoran et al., *Phys. Rev. B*, 55 (1997), pp. R16057–R16060.
40. K. Durst et al., *Acta Mater.,* 54 (2006), pp. 2547–2555.
41. J.G. Swadener et al., *J. Mater. Res.*, 16 (2001), pp. 2091–2102.
42. L.E. Seitzman, *J. Mater. Res.*, 13 (1998), pp. 2936–2944.
43. J. Thurn and R.F. Cook, *J. Mater. Res.,* 17 (2002), pp. 1143–1146.
44. L. Ma and L.E. Levine, *J. Mater. Res.*, 22 (2007), pp. 1656–1661.
45. L. Ma et al., *J. Mater. Res.* (in press, March 2009).
46. J.K. Mason, A.C. Lund, and C.A. Schuh, *Phys. Rev B*, 73 (2006), p. 054102.
47. D.F. Bahr, D.E. Wilson, and D.A. Crowson, *J. Mater. Res.,* 14 (1999), pp. 2269–2275.
48. D.F. Bahr and G. Vasquez, *J. Mater. Res.*, 20 (2005), pp. 1947–1951.
49. M.M. Biener et al., *Phys. Rev. B*, 76 (2007), p. 165422.
50. S. Bhagavat and I. Kao, *Mater. Sci. Eng. A*, 393 (2005), pp. 327–331.
51. K.A. Nibur, D.F. Bahr, and B.P. Somerday, *Acta Mater.*, 54 (2006), pp. 2677–2684.
52. A. Barnoush and H. Vehoff, *Scripta Mater.*, 58 (2008), pp. 747–750.
53. J.A. Zimmerman et al., *Phys. Rev. Lett.*, 87 (2001), p. 165507.
54. A. Hasnaoui, P.M. Derlet, and H. Van Swygenhoven, *Acta Mater.*, 52 (2004), pp. 2251–2258.

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