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# Research Report

## Yielding mechanics on the nanometer scale: What makes it different from the macroscopic world?

U. Dürig and A. Stalder*

IBM Research Division
Zurich Research Laboratory
8803 Rüschlikon
Switzerland

*Present address: Physics Dept., McGill University, Montreal H3A 2T8, Canada



IBM Research Division
Almaden · T.J. Watson · Tokyo · Zurich

# Yielding mechanics on the nanometer scale: What makes it different from the macroscopic world?

U. Dürig and A. Stalder*

*IBM Research Division, Zurich Research Laboratory, 8803 Rüschlikon, Switzerland*

**Abstract:** It has been known for some time that the resistance to plastic deformation increases as dimensions approach the submicrometer scale. The most dramatic effect has been discovered most recently in nanometer-sized Au contacts whose ultimate strengths are comparable to those of the hardest metallic materials known. We show in this Letter that topological material hardening is a general phenomenon originating from the one-dimensional nature of dislocations. Consequently, the corresponding mechanical stresses scale inversely with dimension. At a length scale of typically 10 nm the stresses become comparable to the intrinsic lattice strength. Below this crossover point, dislocations are suppressed completely, and plastic deformation proceeds by a collective slip of entire atomic planes and ultimately by order–disorder transitions. Topological hardening has far-reaching implications, particularly in the context of nanostructured materials.

*Present address: Physics Dept., McGill University, Montreal H3A 2T8, Canada

Metallic contacts have traditionally been studied in indentation experiments [1–4]. Spurred by the development of scanning tunneling microscopy, these experiments have been extended to the nanometer regime [5,6], where point contacts of atomic dimensions have been made [7–11]. A common feature of all these experiments is the observation of a jump to contact involving the spontaneous formation of a nanometer–sized neck as the tip approaches the surface. Such connective necks provide an ideal test ground for studying nano–mechanical material properties.

Major contributions to the field of nano–mechanics have been made with molecular dynamics simulations [12,13]. In particular, Landman et al. [14] were the first to investigate narrow metallic adhesion necks of nanometer size. They found that elongation and thinning of such necks proceeds in a repetitive sequence of mechanical instabilities involving either structural transformations between ordered and disordered states of a narrow region within the neck, or collective shear relaxations of entire atomic planes along crystallographic glide planes [15]. However, no evidence was found of the creation and subsequent propagation of dislocations that would correspond to the classic picture of plastic deformation in metals. Moreover, extremely high values of the order of 5 GPa were obtained for the critical yield stress of Au necks. This value is remarkable because Au is known to be highly ductile on a macroscopic scale, which is reflected by a low value of the order of 200 MPa for the corresponding yield strength.

Indeed, experiments performed on Au necks have confirmed the simulations [16,17]. In particular, yield strengths of the order of 5 to 10 GPa have been measured in necks with nanometer-scale diameters. In addition, we found in our experiments that the yielding behavior and, correspondingly, the contact strength are size-dependent. Contacts were formed by indenting a chemically etched polycrystalline Au tip in a (111)-oriented Au substrate by 10 nm. After indentation, the tip was retracted at a rate of 2 nm/s until the contact broke. The tip–sample applied force was measured using a stiff sensor [18], and the contact area was determined from the measured conductance assuming ballistic quantum transport [19].

By their very nature, indentation experiments are not strictly reproducible. Nevertheless, characteristic features can be identified. Irrespective of the details of the indentation process, such as maximum loading force or contact area, two consecutive yielding phases can be identified when the tip is retracted. Initially the contact diameter remains practically constant until the uniaxial tensile stress exceeds a critical value of the order of 15 to 20 GPa. At this point, the neck yields abruptly. In the first phase, the neck contracts discontinuously in a cascade of violent mechanical instabilities. Slip events in which atomic planes are mutually displaced by sudden collective shear relaxations provide a likely interpretation of these instabilities. A plot of the tensile force versus the contact area at the onset of the instabilities is shown in Fig. 1. The data points cluster around a straight (dashed) line with a slope of $\approx$25 GPa. The prominent feature in Fig. 1 is the offset of 12 nm$^2$ at which the fitted line intersects the horizontal axis, which suggests that necks with a radius of less than 2 nm lose their resistance to plastic deformation entirely. This, of course, is not really the case, but there is a transition to a second yielding phase at approximately this point. Necks that, after elongation, have reached a contact radius smaller than a critical value of the order of 1.5 nm now contract quasicontinuously, whereby the uniaxial yield stress is constant between 6 and 9 GPa. An example is shown in Fig. 1 (solid line: measured data; dotted line: fit to the data with a slope of 8.3 GPa). Moreover, the contact radius shrinks exponentially in small steps each time the neck is elongated by approximately one lattice constant. The exponential decay implies that yielding occurs locally within a narrow region of the order of 1 nm around the constriction of the neck. Furthermore, the energy dissipated in each of these small elongation steps is of the order of 0.1 eV per atom, comparable to the heat of fusion. This suggests that order–disorder

transformations are the apt yielding mechanism [20].
Plastic deformation of necks apparently proceeds by two distinctive mechanisms occurring on a characteristic length scale. How can this be rationalized? For a gedanken experiment, let the constriction of the neck be represented by a cylinder of radius $R$ and height $L$. Compare the energies involved in the following deformation processes: (a) propagation of an edge dislocation [21] along a glide plane perpendicular to the cylinder axis as representative of the classic scenario, (b) mutual displacement by a collective slip motion of two subsections obtained by cutting the cylinder along the glide plane, and (c), motivated by the simulation results, destruction of the crystalline order in the cylinder. These processes are fundamentally different in the sense that respective energies scale as the diameter, the cross section, and the volume of the cylinder. In case (a) we consider only the elastic strain energy, which is maximum when the dislocation line traverses the center of the cylinder. Explicitly one has [22]

$$U_{\mathrm{DISL}} \simeq C\frac{Gb^2}{2\pi(1-\nu)}R\log\frac{R}{R_0} \tag{1a}$$

$$U_{\mathrm{SLIP}} \simeq \frac{Gb}{2}R^2 \tag{1b}$$

$$U_{\mathrm{O-DISO}} \simeq \Delta H_m \pi R^2 L \ \ , \tag{1c}$$

where $G = 45$ GPa (=280 eV/nm$^3$) is the shear modulus, $\nu$=0.38 is the Poisson number, $b = 0.29$ nm, taken equal to the lattice constant of the primitive cubic lattice, is the Burgers vector (1a) or the slip distance (1b), $R_0$=0.29 nm is a cutoff parameter of the order of the lattice constant, $C \simeq 2$ is a finite size correction factor accounting for the interaction of the dislocation line with its image, and $\Delta H_m \approx 6$ eV/nm$^3$ is the heat of fusion which can be taken as a measure of the energy required to create disorder. A plot of the respective energies versus radius is shown in Fig. 2*a*. In the subsequent discussion $L = 2R$ is assumed, corresponding to an aspect ratio of the cylinder (length divided by diameter) of 1. Invoking a minimum energy principle, one concludes from the graph that transitions between yielding modes are indeed expected to occur at a length scale of typically 1 nm.
It is not sufficient to consider the energies alone because the stresses involved can be equally decisive in terms of selecting a particular process. The critical shear stresses for processes (a) and (b) are obtained by taking the derivative of the corresponding energies with respect to $b$ and dividing the resulting shear force by the cross section of the cylinder. Hence, it follows that the critical stress associated with the creation of a dislocation scales as $1/R$, whereas it is constant for the slip deformation (see Fig. 2*b*), and correspondingly one finds a crossover point at $R \simeq 4$ nm. Proceeding with our gedanken experiment, let us subject the cylinder to a uniform shear stress $\tau$. If the radius of the cylinder is larger than 4 nm we expect dislocations to be nucleated as soon as the applied shear stress exceeds the corresponding critical value. For radii smaller than 4 nm the critical shear stress for slip deformations is the lowest, and the yielding of the cylinder will hence proceed by this process even though it is energetically less favorable for $R > 1.5$ nm than for the nucleation of a dislocation line. For radii smaller than 0.85 nm, yielding by order–disorder transitions eventually becomes the energetically most favorable process.
The mechanical energy stored in the neck must exceed the corresponding activation energy in order for any of the processes to occur. For the isolated cylinder the condition reads

$$\frac{1}{2}\frac{\tau^2}{G}\pi R^2 L > U \quad , \tag{2}$$

where $U$ is given by Eq. (1). This condition turns out to be fulfilled for all but very small radii. Thus, the energy requirement affects the critical stress basically only for the order–disorder yielding process. Combining Eqs. (1c) and (2), one obtains $\tau_c \simeq (2\Delta H_m G)^{1/2} \simeq 9$ GPa. In reality the cylindrical constriction is mechanically coupled with the remainder of the neck. Accordingly, the stored elastic energy is larger by a factor of the order of $\int R^2/(LR^2(z))dz$, where $R(z)$ is the radius of the neck. This factor is typically rather large for long and slender necks and of the order of 1 for short and highly constricted necks. Thus, $\tau_c = 9$ GPa refers to an upper limit, whereas the critical yield strength for slip deformations provides a logical lower bound because this process also involves the breaking of bonds.

The principal experimentally observed features can be understood within this framework. However, note that the measured yield stresses are significantly greater than the critical stresses one would expect. This is because the decisive quantity is the resolved shear stress, which is always smaller than the uniaxial stress (it is assumed that radii of curvature are comparable to or larger than the contact radius). As expected, the discrepancy becomes smaller for very short and narrow necks in which the stress distribution is less uniform.

According to our reasoning, finite-size hardening should also manifest itself in the classic yielding scenario in which plastic deformation is tied to the motion of dislocations. The phenomenon is thus not restricted to extreme situations but it is a universal material property originating from the fact that the mean energy density associated with the elastic strain field increases with decreasing structure size. The hardening should become noticeable when the critical shear stress due to the elastic strain field surpasses the intrinsic barrier for dislocation motion. In single crystals this barrier is given by the Peierls–Nabarro stress. The latter is approximately 4 orders of magnitude smaller than the critical shear stress for slip and, hence, finite size effects are expected to become manifest already on the micrometer scale. On the very small scale, there is first a transition from the dislocation to the slip deformation regime and the yield stress levels off correspondingly at a constant value of the order of GPa. The suppression of dislocations is expected to occur at a characteristic radius of $\approx$5 nm irrespective of the material. This follows from the linear $G$ dependence of the critical shear stress for both deformation processes. Furthermore, we expect only a weak logarithmic dependence on the aspect ratio in the expression for the elastic energy associated with a dislocation line. On the other hand, the second transition from the slip to the order–disorder regime depends critically on the aspect ratio of the cylinder as a consequence of the different scaling dependences of the respective activation energies. Yielding by order–disorder transformations can occur only in short and narrow constrictions. Finally, one must also include fracture as a possible deformation mechanism. It has been omitted here because it is not relevant for Au. Fracture becomes important, however, for brittle materials such as W or Ir and particularly for Si. The scaling of the activation energy for fracture is similar to that for slip deformations. Hence, the overall picture does not change fundamentally.

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[21] Note that the order of magnitude of plastic energy is the same for all types of dislocations.
[22] See for example Dieter, G. E. in *Mechanical Metallurgy*, Ch. 4 and 5, McGraw–Hill Series in Material Science and Engineering (McGraw–Hill, New York, 1986).

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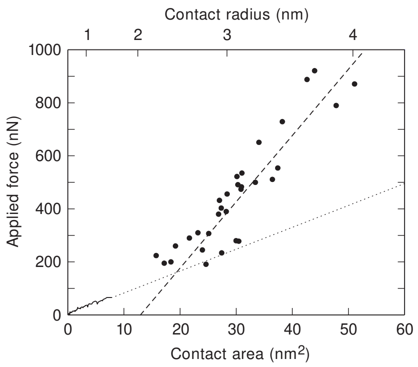


FIG. 1. Applied force versus contact area. Solid dots: Data points recorded at the onset of mechanical slip instabilities during the first yielding phase. Note that the points cluster around a dashed line with a slope of 25 GPa, which intersects the horizontal axis at an offset of 12 $nm^2$. A second yielding phase sets in when the contact radius has become smaller than 1.5 nm. Now the necks contract in a quasicontinuous fashion (solid line) at a constant uniaxial stress between 6 and 9 GPa. The dotted line is a fit to the data with a slope of 8.3 GPa in this particular example.

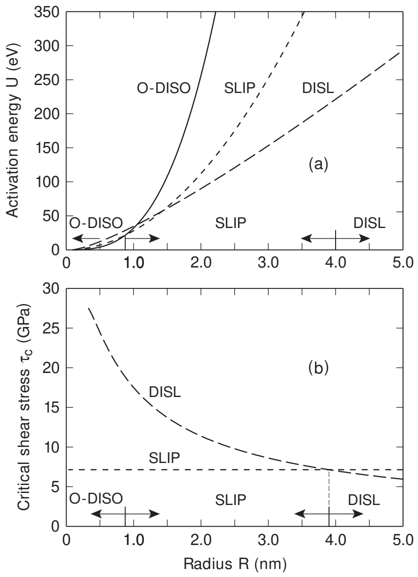


FIG. 2. Considering a cylinder whose height is equal to its diameter, (a) the activation energies and (b) the critical shear stresses are plotted as a function of the cylinder radius for order–disorder transitions (O–DISO, solid line), for collective slip deformations (SLIP, short-dashed line), and for the propagation of edge dislocations (DISL, long-dashed line). Note the three yielding regimes denoted by vertical bars and arrows, each occurring on a characteristic length scale.