Effect of Surface Steps on the Plastic Threshold in Nanoindentation

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Using interfacial force microscopy and passivated Au surfaces, we have investigated the effect of surface steps on the initiation of plastic yield by performing nanoindentations as a function of separation between the probe and neighboring steps. The mean stress at yield was 30%–45% lower at a step than in regions free of surface defects. In addition, the spatial extent of the step's influence was found to be approximately 3 times the contact radius at the yield threshold, suggesting that yield processes are not limited to the region in contact with the indenter. [S0031-9007(98)07614-5]

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Nanometer-scale indentation is now widely applied to measure mechanical properties on a local scale. The technique has proven most valuable in quantifying the properties of small-scale structures such as thin films. However, because of the inherent nature of nanoscale indentation, it also provides an unprecedented opportunity to understand the physics and chemistry of how local structure is linked to observed mechanical properties. Though progress has been made, no clear picture of this link at the nanometer level has yet been developed. To this end, we have studied a model system with controlled atomic-level structure (i.e., individual steps) in order to determine their influence on detailed mechanical behavior.

It has long been recognized that mechanical properties are controlled by the density of defects that act as sources of heterogeneous nucleation of dislocations. The effects of preexisting dislocations [1], inhomogeneities [2], grain boundaries [3], and surface scratches [4] on mechanical properties have been well documented. Recent nanoindentation studies on well-prepared Au single crystals in regions free of surface defects also give measured yield strengths comparable to the predicted ideal strength of the lattice [5–7]. Surface defects, in the form of surface steps, have been observed to affect the yield stress measured by nanoindentation [7], but the effect of a single surface defect has not been explored. By performing indentation tests in the vicinity of a single surface defect, we have measured its effect on heterogeneous nucleation, and by controlling the position of the probe with respect to the defect we are able to gain some understanding of the processes controlling the onset of plastic yield.

Mechanical measurements were performed using the interfacial force microscope (IFM). This instrument, which has been described in detail elsewhere [8], is distinguished by its use of a novel electrostatically driven, force-feedback sensor to ensure rigid displacement control during a loading experiment. This characteristic of the sensor will be exploited to quantify different sample responses arising from differences in surface morphology. In addition, the IFM enables one to perform constant-force imaging, similar to other scanning force techniques. Imaging at a low force (0.2 μN) allowed us to locate surface steps and to characterize the permanent deformation after indentation.

The force sensor and piezoactuators were calibrated using a standard laboratory electronic balance and a displacement indicator, respectively. Both calibrations are necessary for quantitative measurements. Probes used in this study were electrochemically etched 100 μm tungsten wires with tip radii and a parabolic shape determined by field emission scanning electron microscopy (four probes were used with radii of 250, 720, 1000, and 2300 Å). Single-crystal Au samples with (111), (001), and (110) orientations were Ar-ion sputtered and annealed at 950 °C in cycles until the only observed defects were surface steps. Immediately after cleaning, samples were immersed in a 0.5 mM solution of hexadecanethiol [$CH_3(CH_2)_{15}SH$] in ethanol for 24 h to develop a self-assembled monolayer (SAM) of hexadecanethiol to passivate the probe-sample interaction. Without the SAM, a strong adhesive interaction occurs between the Au and W tip and the material is plastic on contact [9,10]. Passivation eliminates this adhesion [9,11] and allows us to analyze elastic force profiles using the Hertzian theory [12,13], which predicts the elastic behavior of a parabolic tip and planar sample using continuum elasticity. If the applied stress exceeds the elastic limit of Au, plastic deformation is observed through a deviation from the Hertzian response and a hysteresis loop in the loading cycle. With the probe radius (R) known, we can quantify the mean applied stress at yield (σ_y) from the measured force (F) and depth of deformation (δ) using the relationship [12],

$$\sigma_y = \frac{F_y}{\pi R \delta_y}, \tag{1}$$

where the subscripts refer to values at the yield threshold.

A typical portion of the surface of a specimen is shown in Fig. 1(a) in a repulsive-force image covering an area 7500 Å × 7500 Å. Two steps can be seen: a single step having a height of 2.5 Å and a double step with a 5.0 Å high step. Intrastep separations within multistep defects were smaller than the lateral resolution of the

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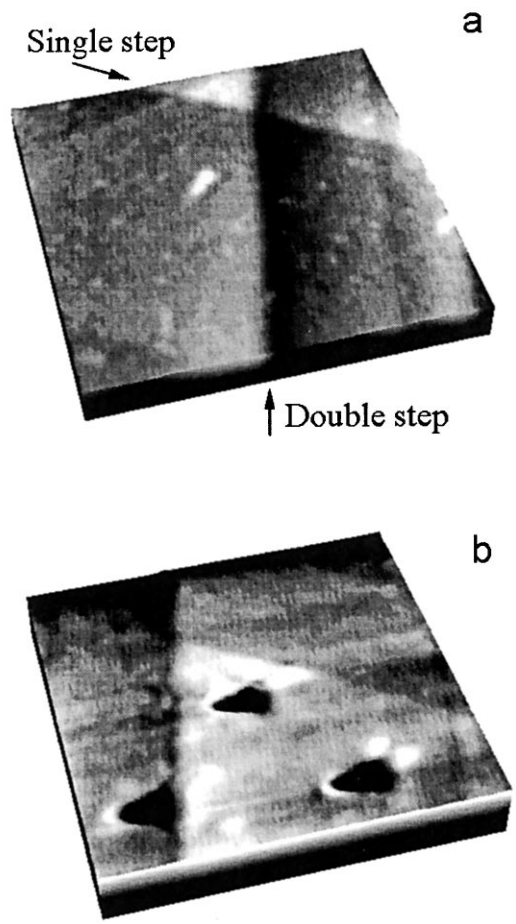

FIG. 1. (a) Constant-force 7500 Å × 7500 Å image of the Au (111) surface depicting a monatomic step (2.5 Å high) and a double step (5 Å high). The double step consists of two monatomic steps with a separation smaller than the lateral resolution of the probe (≈100 Å). (b) This shows the same surface (different location) after several indentations.

imaging technique (in this case ≈100 Å), such that step bunches appeared as single step edges. The resolution also precluded the observation of smaller defects, such as adatom or vacancy islands with diameters <100 Å. Loading data were acquired at positions that varied in distance to the nearest step, as illustrated in Fig. 1(b).

As a demonstration of the effect of surface defects on loading cycles, Fig. 2 presents results from indentations acquired far from and near to a step edge. The force was recorded at displacement increments of 0.5 Å, and in both cycles, the force rises nonlinearly with the depth of deformation following the Hertzian relationship until plasticity begins. The deviation from Hertzian behavior, indicated by the open arrows in Fig. 2, takes the form of relaxation "events" in which the load drops suddenly to some percentage of its threshold value. Loading was continued until the repulsive force reached 21.5 μN, at which point the probe was retracted. The loading cycle of Fig. 2(a) was obtained with the indenter centered between the edges of a terrace that was wider than 5000 Å (far from a step). In this case, yield occurred at approximately ~19 μN. The load drop during the initial yield event was nearly 100%, and further plasticity also occurred in smaller, but significant, relaxation events. The depth of the residual indentation was 150 Å. In Fig. 2(b), the indenter was positioned on a 1550-Å-wide terrace with the indenter axis approximately 300 Å from the 28-Å-high step edge. In this case, the yield event occurred at a force of ~10 μN, which corresponds to a reduction in σ_y by almost 50%. The load relaxation was only a few percent, and further plasticity occurred through a similar series of smaller events. The depth of the residual indentation was 210 Å.

To quantify the influence of a single defect, threshold yield-stress values near straight, isolated steps were measured on Au (111) surfaces. From a series of loading cycles performed using probes of various radii at various distances from steps of various heights, σ_y was calculated using Eq. (1), the results of which are presented in Fig. 3. A positive distance from the step edge (d) corresponds to indenting on the high side of the step, and negative values correspond to the low side. An error bar is shown on one of the data points for comparison of a typical contact diameter at yield ($2a_y$) as estimated from the relation [12]:

$$a_y = \sqrt{R\delta_y}\,. \tag{2}$$

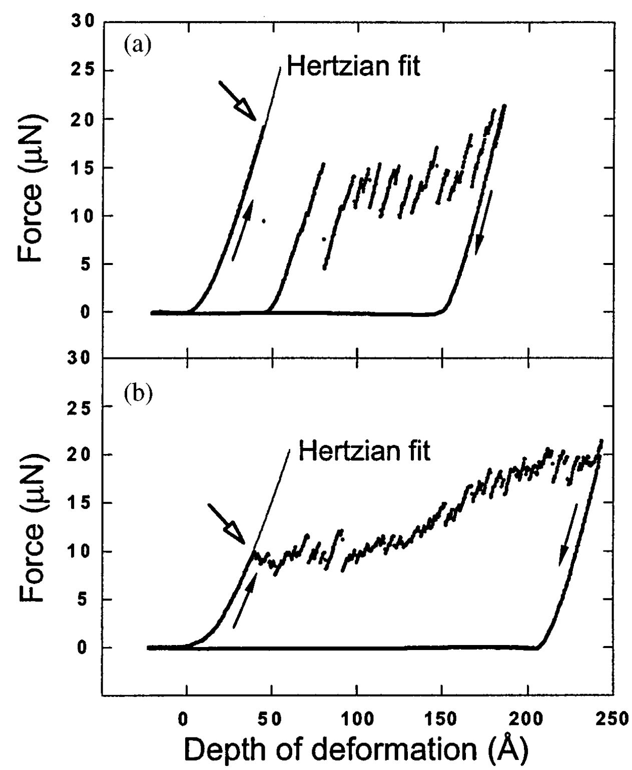

FIG. 2. Indentation loading cycle from Au (111) demonstrating the effect of steps on plasticity. Open arrows identify yield thresholds. (a) Loading cycle when the terrace width is greater than 5000 Å and indenter is centered between the terrace edges. (b) Loading cycle when the indenter is on a 1550-Å-wide terrace, 300 Å from one terrace edge.

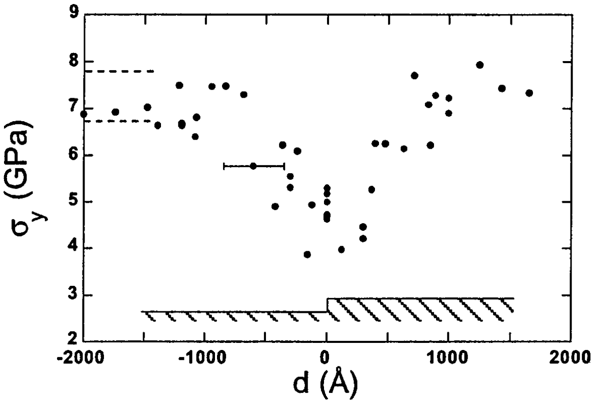

FIG. 3. Measured mean stress at yield as a function of step-indenter separation d for steps on the Au (111) surface having heights ranging from 5 to 30 Å. Positive d values indicate that the indenter is on the high side of the step, while negative values refer to the low side. An error bar is shown on one data point to represent typical diameters of the contact area at the yield threshold relative to the probe/step separation.

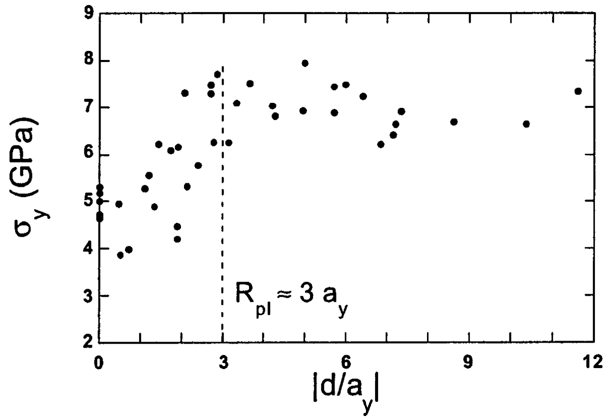

FIG. 4. Measured mean stress at yield as a function of $|d/a_y|$ (absolute value of distance from the step normalized by elastic contact radius prior to yield). When $|d/a_y| < 2$, the yield stress is reduced by a neighboring step, while for $|d/a_y| > 3$, the yield stress is unaffected. The average size of the plastic zone (R_{pl}) for a surface without defects is indicated by the dashed line.

The horizontal broken lines on the upper portion of the ordinate bound the range of σ_y found for Au (111) on wide, defect-free terraces [5]. When the indenter is positioned directly above the edge of the step ($d = 0$), σ_y is only 60%–70% of the σ_y found on the defect-free terraces. It is important to point out that a decrease in σ_y is observed not only when the contact area and step overlap, but even when the two are considerably separated. This indicates that there is some interaction between the step and the stress field outside of the contact area. Note that whether the indenter was on the low or high side of a step made virtually no difference.

To quantify the spatial dependence of the reduction in σ_y, we have replotted the data of Fig. 3 in Fig. 4 as a function of $|d/a_y|$ instead of d. Whenever $|d/a_y| < \sim 3$, lower σ_y values were observed, and whenever $|d/a_y| > \sim 3$, σ_y was no different from that found on the wide, defect-free terraces. The transition between the two regimes is not distinct and occurs somewhere in the range of $2 < |d/a_y| < 3$. These results suggest that a step does not affect σ_y when it is a distance greater than $\sim 3a_y$ away from the indenter, but that it *does not* have to be within the contact area to affect plastic yield.

The spatial extent of plasticity was quantified by measuring the dimensions of the pileup resulting from indentations on defect-free terraces. In an effort to isolate the plasticity due to the initial yield event, loading was reversed immediately after the instrument identified a drop in load of 10% within any displacement increment of 0.5 Å. From 20 topographs, the mean periphery of the pileup, identified as the extent of the region disturbed by the indentation, was found to be at a radius of $\sim 3.2a_y$ (with a standard deviation of 0.45). Taking the radius of the pileup to be the plastic zone radius (R_{pl}) we find that $R_{\mathrm{pl}} \approx 3.2a_y$, which is in agreement with previous nanoindentation studies [14,15].

The processes involved in initiating yield include the nucleation of dislocation loops occurring at the position where the shear stresses are maximum [6,7,16]. The point of maximum shear is below the probe tip and well within the contact area, yet we see that a defect at some distance outside the contact area can reduce σ_y significantly. Based on continuum mechanics [12], stresses outside the contact area fall off as $(a_y/r)^2$ and are principally tensile in nature. Thus, shear stresses at $r = 3a_y$ can be expected to be more than an order of magnitude lower than the maximum values. Still, dislocation nucleation could be precipitated by the step's presence if the step acts as a significant stress concentrator. The continuum-level stress concentration factor is approximately $\sqrt{h/\rho}$, where h is the step height, and ρ is the radius of the step at its base [17]. While we did see a scaling effect with h, to achieve shear-stress levels at the step that equal the shear stresses under the indenter would then require $h/\rho > 100$, meaning that ρ must assume subatomic values. The continuum approach is an inadequate approximation for this case, since a step is by its very nature an atomic-level feature. Paradoxically, the lateral size scale of the effect is orders of magnitude larger than the atomic scale. The solution to the problem, including a full understanding of the detailed processes responsible, will have to await calculations which are able to handle the dimensions of the experiment while including the atomic-level details in the neighborhood of the step boundary.

In summary, we have performed nanoindentation experiments to quantify the effects of surface defects on the initiation of plastic yield. Indenting near a step reduced σ_y by 30%–45% and reduced the magnitude of yield-relaxation events. In addition, we have identified a characteristic length within which surface defects affect indentation processes, and this length is $\approx 3a_y$. These observations indicate that processes that initiate yield are not localized to the point of maximum shear, but can be influenced by the concentration of stress due to inhomogeneities. The results of this study not only identity the effect of surface defects on indentation, such that we have a better idea of how to correlate results from atomically smooth and rough surfaces, but they also give an indication that yield processes are less localized than previously appreciated.

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