Nanoscale fracture studies using the scanning force microscope

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We present a variety of experiments concerning friction and fracture mechanisms using two-dimensional arrays of microfabricated nanotowers as templates. The scanning force microscope tip is used as a tool to apply well-defined forces to the surface of the patterned substrate. Force statistic measurements reveal information about the forces involved in the fracture process and the probability of fracture of selected towers. These methods are discussed in the context of nanometer-scale mechanisms. Using optimized parameters, a controlled removal of individual nanotowers and the ability to “write” predefined patterns on a nanometer scale can be achieved.

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In the past few years many efforts have been made towards controlled surface modification using scanning probe microscopy techniques. Writing patterns in polymers,¹ the modification of semiconductor surfaces, e.g., producing lines^{2,3} and dots,^{4,5} and the positioning of atoms⁶ and molecules,⁷ nanocrystals,⁸ and particles⁹ have been performed successfully. Nanotribological investigations with simultaneous measurements of topography and friction forces can be performed using the scanning force microscope (SFM).¹⁰ The forces involved in the sliding or rolling of nanocrystals⁸ or carbon nanotubes¹¹ on substrates have been examined.

Here, we present a method to perform nanoscale experiments using the scanning force microscope: using lithographically produced nanostructures, fracture and friction are explored at small dimensions.

We used a Topometrix Explorer SFM in contact mode at room temperature. The silicon-nitride tips (Microlevers by Park Scientific Instruments, type A) were calibrated by the method described by Hutter and Bechhoefer¹² obtaining a spring constant of 0.045 N/m. All experiments were performed in bi-distilled water with samples consisting of square SiO₂-nanotower arrays on a silicon substrate produced by laser interference lithography.¹³ The lattice constant is 680 nm and the height of the towers is 120 nm [Fig. 1(a)]. The image in Fig. 1(a) has been achieved by scanning in contact mode with a normal force of 4 nN (“imaging mode”). Moving the SFM tip laterally across one selected tower with an increased normal force results in fracture and removal of this tower [see the center nanotower in Figs. 1(a) and 1(b)]. In a detailed series of experiments, all relevant parameters have been identified and a variety of surface modifications have been observed. Fracture can be achieved by two different modes: In the first mode, the normal force F_N is set to a fixed value of up to 25 nN [“static modification mode,” Fig. 1(c)]; in the second mode the normal force is doubled with respect to the imaging mode and cantilever oscillations with frequencies of 1.75 kHz and amplitudes of 25 nm exert periodic forces in the contact area of the cantilever tip (“dynamic modification mode”).

The involved static and dynamic force magnitudes are the main parameters to control the modification process. The frequency of force modulation was not observed to be of critical importance in the range of 1–3 kHz. In both modification modes the structure breaks at the bottom. The roughness of the residual surface left after removal of the tower is <3 nm. This indicates that the fracture occurs at the SiO₂/Si interface at the bottom of the towers [Fig. 1(c)].

In repeated experiments the statistics of nanotower breaking in the static modification mode has been analyzed. Scanning across $10 \times 10 \mu\text{m}^2$ with an increased force removes a certain fraction of towers; this fraction depends on the force F that is applied to the towers. From the subsequent

FIG. 1. Selective removal of SiO₂ nanotowers using scanning force microscopy. (a) SiO₂ nanotowers with a lattice constant of 680 nm and a height of 120 nm on a Si substrate produced by laser interference lithography. (b) Moving the SFM tip across the center tower in (a) with a normal force F_N of 20 nN results in fracture and displacement. Fracture can be achieved by two different techniques: increasing the normal force (static modification mode) or increasing the normal force and exciting cantilever oscillations additionally (dynamic modification mode). In both cases, the structure breaks at the bottom. The roughness of the residual surface left after removal of the tower is <3 nm. This indicates that the fracture occurs at the SiO₂/Si interface at the bottom of the towers. (c) Diagram of the static modification process. The normal force F_N is increased with respect to the imaging mode to a fixed value of up to 25 nN. The SiO₂/Si interface is denoted by the thin line.

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FIG. 2. Force-dependent statistics of nanotower removal. (a) Scanning across $10 \times 10 \mu\text{m}^2$ in static modification mode removes a certain fraction of towers. From the subsequent overview taken in imaging mode this fraction can be determined. (b) The percentage of removed towers is plotted for a series of such experiments with varied modification force (5–25 nN). No nanotowers have been removed below the observed threshold loading of 13 nN. (c) With repeated static modification mode scanning a monotonously increasing number of nanotowers is removed. Applying 22 nN, the number of removed towers reaches a value of 18% asymptotically. Using only 3 nN more, the surface is completely cleaved after three scans.

overview taken in the imaging mode [Fig. 2(a)], we observe the fraction of the towers that was removed in the previous static modification mode scan. Tower fracture originates from the force F superimposed from lateral and normal forces while scanning the tip across the sample. The coefficient of friction for the investigated system was experimentally determined to be 0.22. From geometric calculations we estimate that the absolute value of the force F acting on the towers is in the same order of magnitude as the normal force F_N . The applied force F is proportional to the measured normal force F_N with given frictional and geometrical parameters. Our estimate is experimentally confirmed by measurements of the lateral forces when breaking some towers. Because of this proportionality ($F \sim F_N$) we use the normal force F_N which is directly accessible to describe the results of our experiments.

FIG. 3. Nanopattern generated using the described dynamic modification method after detailed optimization of all relevant parameters. Individual nanotowers have been directly accessed and removed. The achieved degree of control demonstrates the potential of the presented weak link concept for technological exploitation.

In Fig. 2(b), the percentage of removed towers is plotted for a series of such experiments with varied modification force (5–25 nN). No nanotowers have been removed below the observed threshold loading of 13 nN. Repeated scanning in static modification mode increases the number of removed towers monotonously [Fig. 2(c)]. Applying a force of 22 nN to the surface, the fraction of removed towers reaches a value of 18% asymptotically. Using 25 nN, the surface is completely cleaved after only three scans. Thus, the force difference ΔF of only 3 nN determines whether 18% or 100% of the towers are cleaved. Assuming that this ΔF corresponds to a deviation in tower thickness solely, this thickness deviation can be calculated to 35%. From SFM analysis of the nanotowers we determine the thickness deviation of the individual nanotowers to be below 10%. Therefore, the explanation for the force difference ΔF goes beyond the observed size or width fluctuations, but originates from internal structural variations. Possible mechanisms include the weakening of the interface by strain and chemical agents (liquid environment and etching during manufacturing). Recently, dislocation defects at the SiO_2/Si interface in silicon-on-insulator (SOI) wafers have been observed to induce strain.¹⁴ Our considerations are also supported by investigations of chemical interactions with intrusive water at fracture interfaces in mica.¹⁵ These explorations show that a reduced interface energy in water results in a decreasing force required for initiation of brittle fracture.¹⁵ Using the described microfabricated nanotowers with well-defined material interfaces, SFM measurements of normal and lateral forces enable conclusive experiments about fracture in response to applied forces. The initiation and progress of fracture on the nanometer scale and down to atomic or molecular size are also of significant importance for macroscopic behavior.

Using a combination of imaging mode and dynamic modification mode, a controlled nanotower removal technique is achieved. After optimization of all relevant parameters in dynamic modification mode, individual nanotowers can be directly accessed and removed. Following each sequence of modification experiments, the site of removed towers and the progressively evolving pattern is imaged. Using this technique nanometer scale patterns can be generated. As an example of a complex pattern, we show the logo of our laboratory in Fig. 3. These results suggest the feasibility of the write once, read many times (WORM) type of data storage when using nanotower arrays with the stressed

SiO₂/Si interface as a well-defined “weak link” for mechanical removal. A variety of methods (e.g., masked x-ray lithography) allows the production of high-density nanotower arrays for increased storage densities.

To conclude, we have presented investigations on fracture of nanotowers with an SFM tip. Force statistics reveal a very sharp threshold force for fracture initiation. Combining imaging and dynamic modification modes, the controlled removal of selected nanotowers has been achieved. Using this technique complex nanopatterns have been generated. From the observed qualitative and quantitative behavior reported here, we can foresee a number of experiments to elaborate tribological concepts on the nanometer scale. Most exciting is the ability to apply well-defined forces to well-characterized nanometer-sized structures with varying geometrical and structural parameters. The transition from the elastic and plastic behavior at large dimensions down to the atomic and molecular scale provides valuable insight into size effects as observed for electronic and optical properties.^{16,17} Possibly the closest mechanical analogue is the increase of hardness at small loads as has been observed as a result of surface tensions and strains with nanoindenter experimentation and simulation.^{18,19} Finally, the improved understanding of fracture on the nanometer scale may lead to concepts of data storage using engineered mechanical weak links within nanometer-scale structures.

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- ¹T. A. Jung, A. Moser, H. J. Hug, D. Brodbeck, R. Hofer, H. R. Hidber, and U. D. Schwarz, *Ultramicroscopy* **42-44**, 1446 (1992).
- ²C. K. Hyon, S. C. Choi, S. W. Hwang, D. Ahn, Y. Kim, and E. K. Kim, *Appl. Phys. Lett.* **75**, 292 (1999).
- ³R. Magno and B. R. Bennett, *Appl. Phys. Lett.* **70**, 1855 (1997).
- ⁴M. Wendel, S. Kühn, H. Lorenz, J. P. Kotthaus, and M. Holland, *Appl. Phys. Lett.* **65**, 1775 (1994).
- ⁵J. Cortes Rosa, M. Wendel, H. Lorenz, J. P. Kotthaus, M. Thomas, and H. Kroemer, *Appl. Phys. Lett.* **73**, 2684 (1998).
- ⁶D. M. Eigler and E. K. Schweizer, *Nature (London)* **344**, 524 (1990).
- ⁷T. A. Jung, R. R. Schlittler, J. K. Gimzewski, H. Tang, and C. Joachim, *Science* **271**, 181 (1996).
- ⁸P. E. Sheehan and C. M. Lieber, *Science* **272**, 1158 (1996).
- ⁹T. Junno, K. Deppert, L. Montelius, and L. Samuelson, *Appl. Phys. Lett.* **66**, 3627 (1995).
- ¹⁰R. Lüthi, E. Meyer, H. Haefke, L. Howald, W. Gutmannsbauer, and H.-J. Güntherodt, *Science* **266**, 1979 (1994).
- ¹¹M. R. Falvo, R. M. Taylor II, A. Helser, V. Chi, F. P. Brooks, Jr., S. Washburn, and R. Superfine, *Nature (London)* **397**, 236 (1999).
- ¹²J. L. Hutter and J. Bechhoefer, *Rev. Sci. Instrum.* **64**, 1868 (1993).
- ¹³M. Schildenberger, Y. Bonetti, M. Aeschlimann, L. Scandella, J. Gobrecht, and R. Prins, *Catal. Lett.* **56**, 1 (1998).
- ¹⁴P. Sutter, M. G. Lagally, *Phys. Rev. Lett.* **82**, 1490 (1999).
- ¹⁵B. Lawn, in *Fundamentals of Friction: Macroscopic and Microscopic Processes*, edited by I. L. Singer and H. M. Pollock (Kluwer Academic, Dordrecht, Netherlands, 1992), p. 137.
- ¹⁶F. J. Himpsel, T. A. Jung, and P. F. Seidler, *IBM J. Res. Dev.* **42**, 33 (1998).
- ¹⁷M. V. Rama Krishna and R. A. Friesner, *Phys. Rev. Lett.* **67**, 629 (1991).
- ¹⁸R. Komanduri, N. Chandrasekaran, and L. M. Raff, *Phys. Rev. B* **61**, 14007 (2000).
- ¹⁹W. C. Oliver, R. Hutchings, and J. B. Pethica, in *Microindentation Techniques in Materials Science and Engineering*, edited by P. J. Blau and B. R. Lawn (International Metallographic Society, Philadelphia, PA, 1985), p. 91.