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Nanolithography of silicon: An approach for investigating tip-surface interactions during writing

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We report an approach for monitoring tip-surface interactions. The approach is based on power spectrum analysis of atomic force microscope (AFM) tip oscillations during scanning probe nanolithography on Si surfaces. A single mode harmonic oscillator model allows us to determine the main characteristics of the AFM tip-surface interaction: the amplitude of oscillation, the resonant frequency, and the damping factor, during the writing process. We measure these quantities in scanning probe anodization versus the bias voltage. By fixing the length of lithographically patterned lines, and the energy deposited into each line, we search for trends which may reveal the major factors controlling the quality of AFM-written nanostructures. Our data are consistent with the concepts that a water meniscus and electrostatic tip-surface interactions dominate contact AFM lithography. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1413736]

Scanning probe nanolithography is a powerful and very promising technique for modifying the surfaces of semiconductors, metals, and polymer films. Presently this technology in conjunction with atomic force microscopy (AFM) may be divided into two main categories: contact and noncontact (or intermittent contact) mode lithography. The original technique was suggested and implemented by Dagata *et al.*[1] for oxidizing hydrogen-passivated silicon by means of a scanning tunneling microscope (STM). The technique was later extended to AFM contact mode with dc applied voltages by Snow *et al.*[2] and then improved using noncontact mode dc AFM by Perez-Murano *et al.*[3] and Garcia *et al.*[4] The disadvantage of traditional dc AFM lithography is the formation of space-charge fields during oxidation. The negatively charged hydroxyl ions transported through a water meniscus to the surface create a negative space-charge layer which prevents further oxidation and thus vertical growth of the oxide features, which in turn leads to a degradation in their aspect ratio. With this in mind, the goal of this letter is to present ideas and methods that may enhance our understanding of tip-surface interactions during AFM-assisted nanolithography. We demonstrate that the dynamical behavior of the tip under the influence of an applied dc voltage can be used to probe the physics of the oxidation process.

Applied voltage is the simplest parameter to control in scanning probe oxidation. It has been shown that a meniscus of water surrounding the AFM tip is necessary for scanning probe lithography in air,[4] and it is possible that the shape and size of the meniscus may be influenced by the applied voltage. By controlling the voltage we also modify the associated electric field and thus the energy of the ions resulting from the dissociation of water. Several studies have introduced the concepts of measuring the current or maintaining a constant flux or charge per unit length during scanning probe surface oxidation.[5–8] These approaches effectively treat the oxidation process as being independent of the energy (kinetic or electronic) of the ions, or in other words assume that the ions are not involved in multiple scattering events which may alter the kinetics of the reaction by energy redistribution. Since we are interested in studying the voltage dependence of the parameters controlling the writing process, we introduce a different approach: constant energy density (energy/length). Since the energy density per unit time is related to the squared magnitude of the electric field, and this in turn is proportional to the voltage squared, we maintain the product V^2t constant during writing, where V is the voltage, and t is the total time it takes to write a line of fixed length at constant writing speed.

In the work presented here, *p*-type silicon {100} substrates are cleaned in 40 wt% hydrofluoric acid solution for 1–2 min followed by standard AFM imaging to verify the absence of large-scale surface defects. The tip holder is electrically isolated from the rest of the AFM, and we employ 200-μm-long silicon nitride cantilevers coated with aluminum by thermal evaporation in a vacuum chamber. The natural resonant frequency of these tips in air is about 5 kHz. A dc power supply is used to apply a voltage between the AFM tip and substrate with the tip biased negatively. The applied energy density is kept constant by adjusting the total writing time of the fixed-length lines in accordance with the applied voltage. Since the writing is done at constant scanning speed in order to minimize hydrodynamic differences,[9] writing at lower voltages (e.g., longer times) is accomplished by scanning over the same line many more times than is necessary at higher voltages. It is possible to use voltages up to 25 V and times as short as 4 s and still acquire enough data for the power spectrum analysis described below.

It is expected that even under static (not scanning) conditions, capillary and electrostatic forces control the attractive nature of the tip-surface interaction. We first probe the influence of the voltage by measuring the snap-on force as a function of voltage under ambient humidity conditions without scanning the tip. Figure 1 clearly indicates that the capillary forces dominate at low voltages and that the net force increases exponentially demonstrating the major role that

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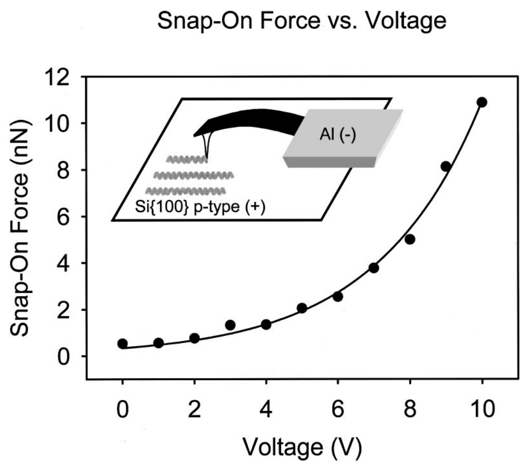

FIG. 1. Snap-on force as a function of voltage. Circles: experimental data. Solid line: exponential fit. Inset: illustration of the writing process to emphasize tip-surface contact.

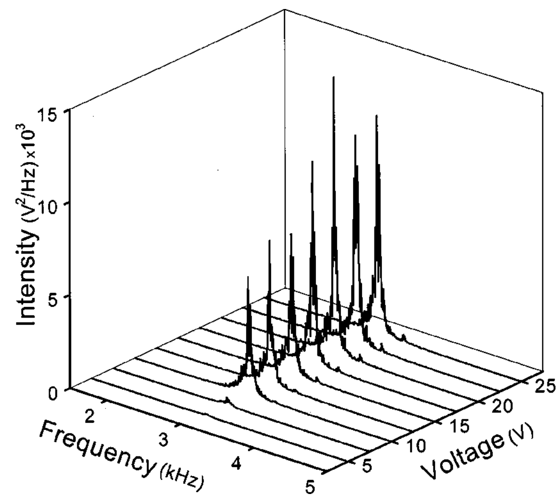

FIG. 3. Power spectra acquired at different voltages in contact AFM lithography on Si{100}. The intensity of tip oscillations varies with voltage with a maximum observed at 17.5 V.

electrostatic interactions play in contact AFM lithography. The solid line is from a numerical fit of a pure exponential function to the experimental data (circles). The inset of Fig. 1 is an illustration of the writing process to be discussed in the remainder of the letter. This is included to emphasize the fact that the tip is bound to the surface during writing, so that the oscillations we measure (via the laser beam that reflects off the back of the cantilever) and analyze during writing are not those of a free cantilever.

A set of silicon-oxide lines written by AFM in dc contact mode at different voltages is shown in Fig. 2. It is obvious that the line profiles depend on the voltage, and that at higher voltages we can write "double lines." The production of double lines has been reported by others,[10] and can be accounted for by considering the implications of Fig. 1. At higher voltages the tip is strongly bound to the surface, penetrating into and altering the shape of the meniscus which allows the "sides" of the tip to participate in ion production. This effect is probably even more pronounced by the fact that at high voltages the tip wears more as we write (since it is more strongly bound), causing it to blunt. The thermal oscillations of the cantilever are recorded during writing using a data acquisition board at 80 kHz. The power spectra of the thermal noise as shown in Fig. 3 are obtained by Fourier transforming the oscillation data using commercial software. Each power spectrum is averaged for a period of 4 s which involves multiple passes over the same line, and often several modes appear in the resulting power spectrum. However, the first mode is dominant over the other modes, and therefore a single-mode harmonic oscillator model is used to fit the data to obtain three major parameters: cantilever amplitude (oscillator strength), damping factor, and resonant frequency. Recalling the inset in Fig. 1, these data represent the vertical flexing or bowing motion of the cantilever during writing with the tip bound by electrostatic forces to the surface because of the applied voltage. Analysis using a single-mode model should therefore be considered as a first approximation toward understanding this complicated dynamical system.

This type of analysis indicates that the oscillation amplitude of the cantilever increases and the damping decreases with an increase in writing voltage, as shown in Fig. 4. It should be realized that an increase in the writing voltage results in a decrease in the total writing time (since we maintain $V^2t=\text{constant}$). Thus lines written at low voltages have been scanned over (at fixed speed) many more times than those written at high voltages. Although no quantitative model of the writing process can be made at present from data sets such as these, the trends (indicated by the lines inserted in Fig. 4 to guide the eye) are reproducible across data collected under different humidity conditions and with different tips and substrates. These trends could be the sig-

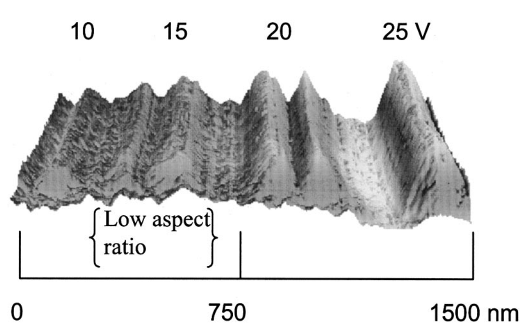

FIG. 2. Silicon-oxide lines written on the surface of Si{100} under conditions of high humidity (60%).

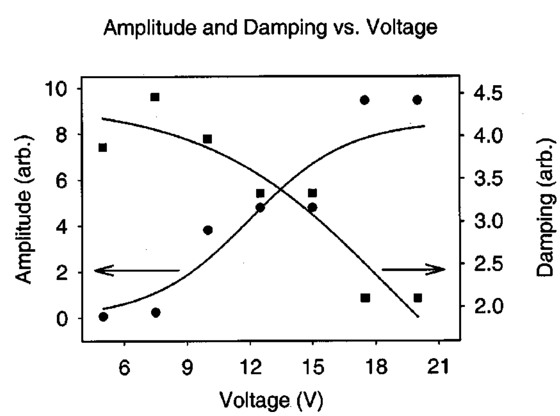

FIG. 4. Amplitude (circles, left axis) and damping (squares, right axis) vs writing voltage extracted from numerical fits to the power spectra recorded during AFM writing on Si in air. Trend lines are inserted to guide the eye.

nature that at higher voltages the shape and size of the meniscus becomes unstable or that multiple-ionization processes and space-charge effects begin to play a more important role. More quantitative modeling of data such as these is in progress and will be reported in a future publication. At present, we feel that our results reported in this letter utilizing an approach (power spectrum analysis) and concept (constant energy density) have the potential to lead us closer to a more complete understanding of the physical processes controlling AFM-assisted nanolithography.

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