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Improving the resolution and the uniformity of AFM tip induced oxide patterns with pulsed voltages

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Abstract

Oxide line patterns were fabricated on the surface of titanium (Ti) film using atomic force microscopy (AFM) tip induced local oxidation technique. The growth behavior of the oxide under static voltages was studied. It was found the lateral growth of the oxide experienced two stages and the growth rate at the initial stage was very high. Pulsed voltages were employed and their effects on the controlling of the oxidation dynamics were examined. The results indicated that the high lateral and vertical growth rates of oxide at the initial stage could be suppressed with pulsed voltages. A minimum line width of 8 nm and highly uniform patterns were obtained with optimized voltage pulses. These results indicated that applying pulsed voltage is an effective method for improving both the resolution and the uniformity of the fabricated structures with scanning probe microscopy (SPM) tip induced local oxidation technique.

1. Introduction

Scanning probe microscopy (SPM) is a high-resolution tool capable of exploring local structures as well as properties of material surfaces on atomic scale [1]. Utilizing local interactions between the SPM probe and the sample surface, SPM can also be used as a powerful lithography tool to modify the local structures or chemical properties of surfaces [2–4]. Among various approaches of SPM based lithography techniques, local oxidation induced by electrically biased SPM tips is proved to be a promising tool for the fabrication of nanometer-scale structures and devices [5,6]. The process of local oxidation of a metallic or semiconducting surface induced by SPM tips is analogous to electrochemical anodic oxidation [7]. In this system, the tip and the sample surface act as the cathode and the anode, respectively, and the water meniscus condensed between the tip and the sample surface from the ambient humidity is the electrolyte. When a negative bias is applied to the tip, the OH^- ions contained in the water meniscus are driven to the sample surface by the electric field and localized anodic oxidation of the sample occurs beneath the tip apex, resulting in the growth of oxide structures with the width of 10–100 nm and height of 1–10 nm.

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Fabrication of novel structures and devices, such as single electron transistor (SET) [8] and etching masks [9], has been demonstrated employing the SPM tip induced local oxidation technique. Although this technique is a versatile and efficient method for fabricating nanometer-scale structures that cannot be achieved by conventional lithography techniques, further improvement of the resolution and reliability of this technique is highly demanded for the fabrication of ultra-fine structures and novel quantum functional devices. However, this remains a great challenge because the mechanism and dynamics of the process of tip induced local oxidation is still under study [10,11].

In this work, oxide line patterns were fabricated on the surface of titanium (Ti) thin films using a conductive atomic force microscopy (AFM). The growth behavior of the oxide lines under static voltages and pulsed voltages was comparably studied to identify the factors that affect the resolution and reliability of the fabricated structures. It was found that controlling the oxidation process at the initial stage was important for the quality of the fabricated patterns. The improvement of both the resolution and the uniformity was achieved with pulsed voltages for their efficient suppression on the high growth rate of oxide at the initial stage.

2. Experimental details

The sample was prepared by magnetron sputtering of 10 nm thick Ti film on the Si(1 0 0) substrate. The sample was ultrasonically cleaned in ethanol and dried in air before patterning. Local oxidation of Ti films was carried out using a commercial Solver P47 AFM (NT-MDT, Russia). The AFM worked in contact mode and the feedback loop was kept on in the process of fabrication. Commercial conductive AFM tips (NT-MDT, Russia), which had a 25 nm coating of WC_2 as the conductive layer, were used in both the oxidation process and subsequent characterization of as-grown oxide patterns. The radius of the tip was less than 35 nm. The relative humidity of the ambient was kept constant at values ranging from 20% to 60% during experiments. To reduce the influence caused by the tip convolution effect, the FWHM (full-width at half-maximum) was measured as the width of oxide lines.
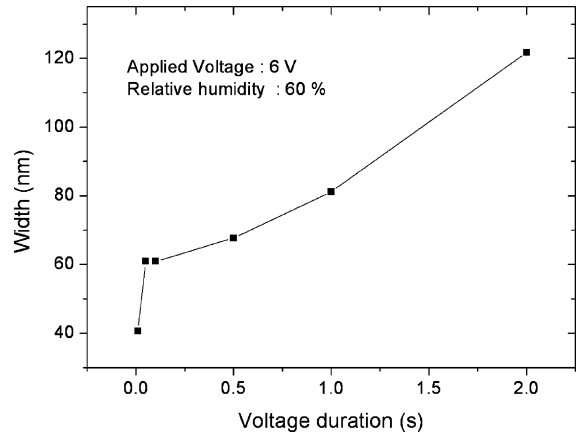

Fig. 1. The dependence of the width of oxide lines vs. the duration of applied voltages.

3. Results and discussion

In order to explore the lateral growth behavior of the oxide, line patterns were fabricated by applying static voltages with different durations. Fig. 1 shows the dependence of the width of the oxide lines versus the voltage duration. The applied voltage was 6 V and the relative humidity was 60%. It can be seen from Fig. 1 that the increase of the oxide width versus the applied voltage duration exhibits two stages. The lateral growth rate of oxide at each stage could be deduced by linearly fitting the width data and the voltage duration. The oxide width increased rapidly with a growth rate of about 507 nm/s when the voltage duration was below 0.05 s. Then the growth rate decreased to about 33 nm/s with the voltage duration longer than 0.05 s. This result can be explained with the non-uniform distribution of the electric field. The applied voltage produced a highly localized electric field between the tip and the surface. Once the voltage was applied to the tip, the OH^- ions drifted to the sample surface under the produced electric field, but their drifting speeds were different spatially and the ions would arrive at the surface at different time. The surface sites under the tip apex were firstly oxidized and then the oxidized sites extended away. Because the electric field was intense and the initial oxidation occurred at the uncovered surface, the oxidation speed was high, which resulted in the observed high lateral growth rate at the initial stage. The diameter of the oxidized region in this stage was comparable with the

neck of the water meniscus. Then the lateral growth rate of oxide would slow down because the electric field strength decreases rapidly outside the central region oxidized initially. On the other hand, the as-grown oxide at the initial stage also reduces the electric field strength and declines the drift speed of OH^- ions to the underlying Ti surface.

The fabricating resolution is mainly dependent on the lateral growth of oxide induced by the electric field. The relatively high lateral growth rate of the oxide at the initial stage limits the further reduction of the pattern width. With a continuous voltage applied, the smallest lateral dimension of fabricated structures is usually limited to about 20–30 nm, which is determined by the extent of condensed water meniscus between the tip and the sample surface. Further improvement of the fabricating resolution needs to control the lateral growth of oxide, which cannot be realized with a continuous voltage. So we examined the growth behavior of oxide with a pulsed voltage applied. Fig. 2 shows the dependence of the width of the patterned lines versus the number of applied voltage pulses (N). The pulsed voltage has a square waveform as shown in the inset of Fig. 2. The pulse amplitude (V1) was set to be 9 V. The effective oxidation time of the pulsed voltage was set to be equal ($T1 \times N = 0.2$ s) for different lines. The rest duration of the pulse (T2) was 50 ms and the corresponding amplitude (V2) was 0 V. So the active duration of the pulse (T1) varied from 0.2 to 0.004 s as the

pulse number increased from 1 to 50. The relative humidity was kept at 46% during the fabrication process. It can be found from Fig. 2 that the variations of the width of oxide lines are remarkable and show a continuous decrease to about 24% with the pulse number N increased from 1 to 50. The reason for the suppression on the oxide width by a pulsed voltage can be understood with the lateral growth character-istics of oxide. As shown in Fig. 1, the growth of oxide in width experienced two stages and the growth rate at the initial stage was relatively high. When a static voltage is applied on the tip, OH^- ions drift to the anode continuously and induce the growth of oxide on the sample surface. In the case of a pulsed voltage applied, the continuous transport of the OH^- ions will be broken in the rest duration (T2) of the voltage pulse and the oxidation process will be stopped. When the next voltage pulse is active, the directional transport of the OH^- ions should be re-initialized because the OH^- ions goes randomly in the rest period of the last voltage pulse. As a result, the oxidation process cannot continue from where it stopped but must restart from the beginning. This process repeats within all the voltage pulses and causes the reduction of the lateral growth rate of oxide. If the active duration of the pulse (T1) is sufficient short and the initial stage of the lateral growth is broken, a strong suppression of the oxide growth in width could be achieved, as seen from Fig. 2, a 8 nm wide oxide line was formed with the pulse number $N = 50$.

Fig. 3 gives the dependence of the height of these oxide lines versus the number of applied voltage pulses. It can be found in Fig. 3 that the height of the oxide lines also decreases following the increase of the pulse number N. It has been demonstrated that the local oxidation of the Si surface induced by the tip voltage is an electric field effect and the height of the oxide has a linear relationship with the voltage ampli-tude and a logarithmic relationship with the voltage duration [10,12] under a static or continuous voltage. We got the same results on the surface of Ti with a static voltage (not shown here). The initial vertical growth rate of oxide is high under the intensive electric field and this makes the distance between the tip and the as-grown oxide reduce rapidly. The subsequent adjustment of the tip vertical position by the AFM feedback loop will usually change the oxidation con-ditions on the next scanning point, hence destroy the
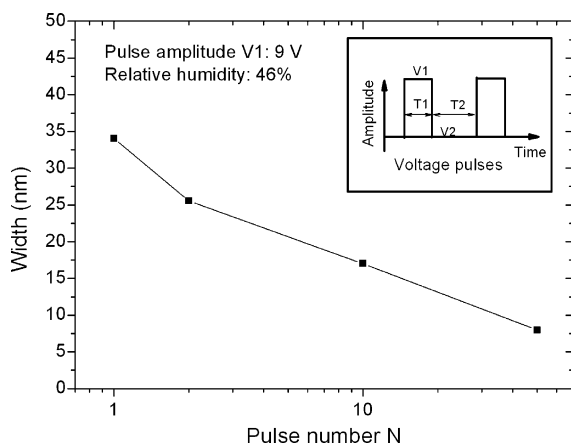

Fig. 2. The dependence of the width of oxide lines vs. the voltage pulse number. The inset shows the waveform and variable parameters of the pulsed voltage.
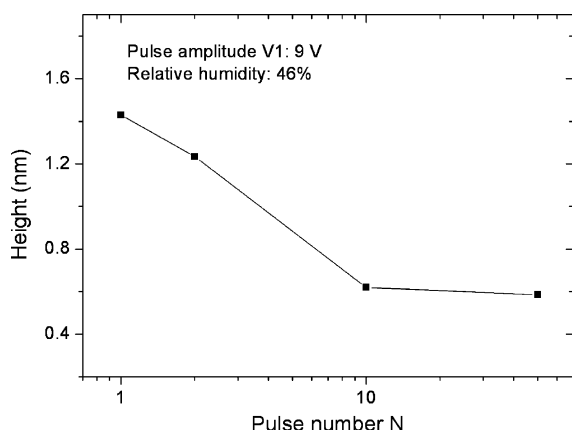
Fig. 3. The dependence of the height of oxide lines vs. the voltage pulse number.

uniformity of patterned structures. Fig. 4 shows a pattern of Chinese characters "Peking University" fabricated by a static voltage with the amplitude of 10 V and the duration of 0.8 s. It can be seen clearly that the structure of the pattern has a poor uniformity and many bright protrusions with the height of 1–4 nm exist on the pattern. As shown in Fig. 3, with a pulsed voltage, the vertical growth rate can also be suppressed by repeatedly breaking the oxidation process. On the

Fig. 4. AFM topographic image of a pattern of Chinese characters formed with a static voltage. Image size: 2 μm × 2 μm.
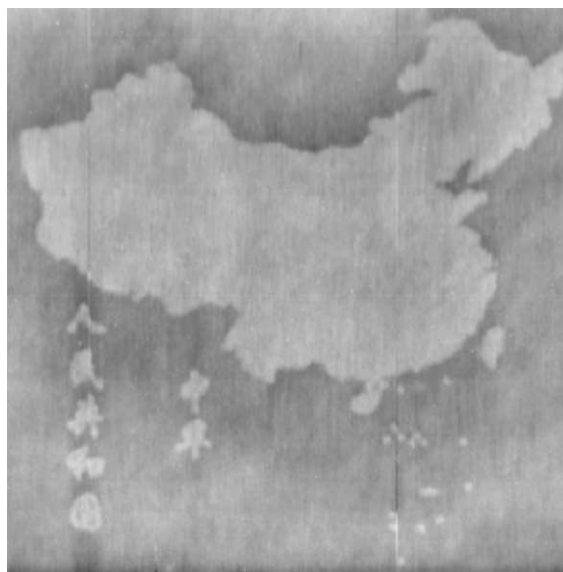

Fig. 5. AFM topographic image of a pattern of the map of China formed with a pulsed voltage. Image size: 2.5 μm × 2.5 μm.

other hand, although the effective oxidation time was equal for different line patterns in Fig. 3, the active duration ($T1$) of each voltage pulse reduced when the number of the voltage pulse was increased, thus changing the duty cycle of the voltage pulse. For $N = 10$, the duty cycle is 40%, and the average amplitude of voltage is only 3.6 V. The reduction of the average amplitude with a pulsed voltage also further decreases the vertical growth rate of oxide, thus improves the uniformity of the fabricated patterns. Fig. 5 shows a pattern of the map of China fabricated by a pulsed voltage with the amplitude of 10 V and pulse number $N = 7$ ($T1 = 4$ ms). No protrusions can be found on the patterned area, indicating that applying pulsed voltages is an efficient method for achieving uniform structures.

4. Conclusions

In summary, the growth behavior of oxide on the surface of Ti films induced by AFM local oxidation with static and pulsed voltages were studied. It was found that the lateral growth of oxide experienced two stages and the growth rate at the initial stage was relatively high. Using pulsed voltages was proved to be an efficient method for suppressing both the lateral

growth rate and the vertical growth rate of oxide at the initial stage. High-resolution and high uniformity can be achieved with an optimal pulsed voltage in the fabrication.

Acknowledgements

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