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EFFECTS OF STM TIP MOVEMENT TO THE REAL WIDTH OF AN ELECTRIC PULSE ACTING ON THE GAP BETWEEN THE TIP AND THE SAMPLE SURFACE

C. Z. Cai, W. L. Wang, X. Y. Chen and Q. Q. Shu

Department of Applied Physics, Chongqing University, Chongqing 400044, P. R. China

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Abstract. During the process of nano-lithography on solid material surfaces by a Scanning Tunneling Microscope (STM) system working at constant current mode, while a high-voltage electric pulse acts on the gap between the tip and the sample surface, the feedback circuit causes the tip to withdraw swiftly, resulting in a reduction of effective pulse-width. In the experiment presented in this paper, a specially designed and computer controlled temporary "holding" function can maintain the tip position and the tip-sample distance during the whole period when the high-voltage pulse acts, so that the effective pulse-width can be accurately measured. An experiment of lithography on a graphite surface in air found, for the first time, the threshold of effective pulse-width to be (0.04±0.01) μS, with a voltage amplitude of 4 V.

1. INTRODUCTION

The scanning tunneling microscope (STM) has been used not only as an instrument for atomic-resolution surface observation, but also as a tool for nanometer-scale surface manipulation [1,2]. The method often used, apart from mechanical indentation with the STM tip, is application of a voltage pulse across the tip and sample, which stimulates some physical and/or chemical processes in the sample surface [3,4]. Because of the sharpness of the tip, one can localize the pulsed electric field within an extremely small neighborhood around the tip, leading to the new technology of so-called nanolithography, or nanometer-scale etching. Although it is not yet realistic to apply this technology directly in practice at this moment, a great number of basic laboratory research work, including attempts of manipulating materials such as Si, GaAs, Ti via different techniques at the atomic or molecular levels [4-9], as well as fabrication of quantum electron devices [9,10], have been done. Nanolithography with either the STM or general scanning probe microscope (SPM) appears to have potential applications in the microelectronic industry.

Many factors, such as the tip-sample distance, the characteristics of the voltage pulse, the properties of tip and the sample materials, the ambient, *etc*., are believed to influence etching results. Some of the factors are relatively easy to control and have been extensively studied. For example, a clear voltage amplitude threshold to write patterns on a highly orientated pyrolytical graphite (HOPG) surface has been observed by different groups. For a given tunneling tip-sample distance, it varies daily from tip to tip and ranges from 3 V to 8 V [11], without obvious polarity dependence [12,13]. Further investigation showed that such a variation essentially comes from the change of humidity, because a well-defined threshold of (4.0±0.2) V was obtained in aqueous solutions [13]. The existence of water molecules also implies that the etching process involves some chemical reactions [12].

However, there seems to be no well-defined threshold in pulsewidth for the etching process [11,14]. According to reports to date, the pulsewidth to write pits in an HOPG surface can be as short as one microsecond [11], or as long as a few hundred microseconds [11,12,14]. A similar situation also exists in the nanometer-scale fabrication by electrochemical deposition, with an even larger variation in pulsewidth [7,15,16]. Such a pulsewidth dependence can not be reasonable: if the voltage pulse supplies the energy necessary for the chemical reactions to occur on the surface, longer pulses should

Corresponding author: C.Z. Cai, e-mail: btccaicz@nus.edu.sg

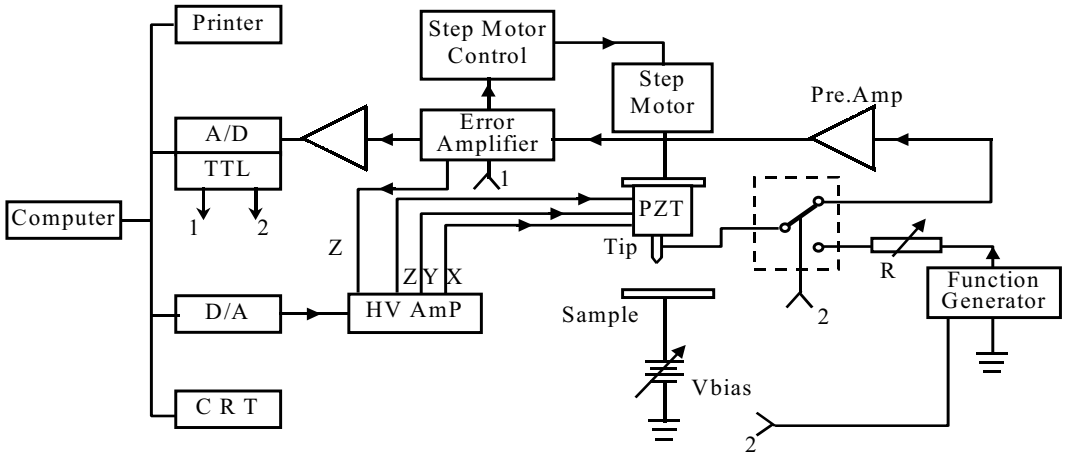

Fig. 1. Block diagram of the STM system used for nanolithography.

simulate more surface atoms to react, resulting in larger pits. A detailed analysis of the reported experiments shows that the pulsewidth independence may come from the tip movement during the voltage pulse. When the STM system operates in the constant-current mode, due to the feedback response a high-voltage pulse applied across the tip and the sample can cause the tip to withdraw swiftly to increase the tip-sample distance. Therefore, the real duration time, *i.e.*, the effective pulsewidth, is much shorter than the applied one. The effective pulsewidth is determined essentially by the system response time, and is independent on the applied pulsewidth within a wide range. In fact, T. R. Albrecht *et al*. and R. H. Bernhardt *et al*. have observed such a withdrawal phenomenon [11,16], and P. Moriarty and G. Hughes have also mentioned that the etching result shows no dependence on the pulsewidth, but is related to the pulse gradient [14].

To investigate the effective pulsewidth and its threshold in nanolithography processes, we modified a home-made STM system by adding a set of hardware and software which can temporarily cut of the feedback loop and hold the tip-sample distance during the etching operation, so that the real effective time of the applied voltage pulse can be measured. With this setup, we obtained for the first time that the threshold of effective pulsewidth to write pits in an HOPG surface in air is about 0.02~0.04 μS.

2. EXPERIMENTAL DETAILS

Our home built STM system is similar to most commercially available STM systems. An additional part for lithography is added, which includes a pulse generator (KIKUSUI-4502 function generator), a hold circuit inserted in the error amplifier and a switch to connect the tip either to the preamplifier input or to the pulse generator output. All three parts are program-controlled. Fig. 1 shows a block diagram of our system. The piezoelectric crystal tube is mounted vertically. The outside thin-film electrode is cut into four equal parts, and two pairs of symmetrical scanning voltages X, $\overline{X}$ and Y, $\overline{Y}$ with a 256x256-steps resolution are applied on the tube. The inside electrode is connected to the error amplifier output to control the tip-sample distance. The sample table is horizontally placed, making it easy to cover the sample surface with liquids if necessary.

To obtain the surface topography of a large area, the system is usually run in the constant-current mode. The tip is connected to the output of the pulse generator and there are two types of procedures for applying voltage pulses to etch. One is "without temporary holding", and the other is "temporary holding". In the former case, the tip withdraws when a voltage pulse is applied due to the feedback response. In the latter case, before triggering a voltage pulse, the feedback loop in the error amplifier is temporarily cut off to hold the tip-sample distance, so that no vertical withdrawal occurs during the whole interval of voltage pulse application. Afterward, the tip is connected back to the preamplifier input, and the error amplifier is turned back to the normal feedback-controlled mode, followed by the subsequent lateral scan. In such a way, the full pulse duration can be considered effective. In the former case,

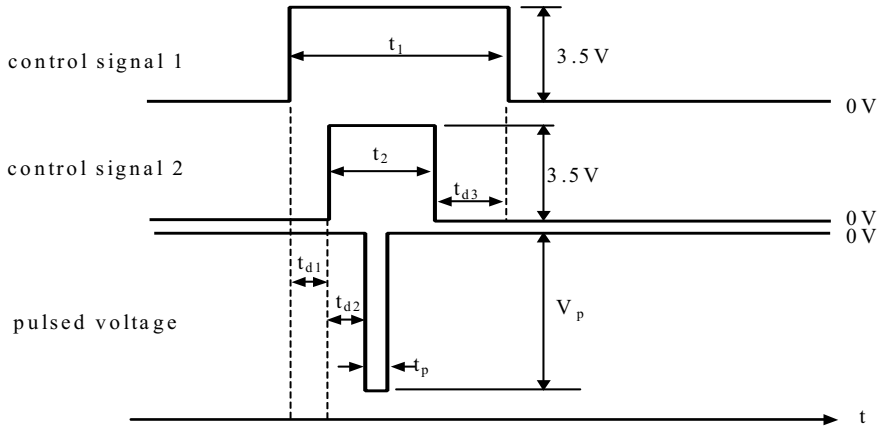

Fig. 2. A typical time order logic of applying a voltage pulse across the tip and the sample.

only a fraction of the pulse duration is effective and the fraction is difficult to measure quantitatively.

A typical time order logic is shown in Fig. 2. Control signals 1 and 2 are also indicated in Fig. 1. When the control signal No.1 is TTL low, the error amplifier works in the normal feedback mode, and when TTL high, the error amplifier takes the "holding" mode. When the control signal No.2 is TTL low, the tip is connected to the preamplifier input, and when TTL high, the tip is connected to the pulse generator output. The durations t_1 and t_2 are computer-controlled, and the pulsed voltage amplitude V_p and the pulsewidth t_p are tuned by the pulse generator. Measured values are typically t_{d1}=7 μS, t_{d2}=5 μS, t_{d3}=10 μS.

There are three ways to control the pattern to write. One is interactive single-point writing. In this way one can move the tip to a selected point on the surface in the scan zone with the help of a moving cursor indicator overlaid on the STM image on the computer display and then send out a command (push a key) to trigger an etching procedure. The second way is multipoint writing. In this way, one can select a set of points in the area of 256x256 points on the computer display and store in memory. While imaging the sample surface, the system stops the tip scanning at each selected point and triggers an etching procedure. The third way is interactive multipoint writing. In this way, while the system scans continuously frame by frame, one can select points following the cursor indicator overlaid on the scanned image and store in memory. Once a set of points is selected, one can start a multipoint etching procedure similar to the second case above. The last method is the most flexible, and is usually used in our experiments.

The HOPG used is obtained from the Metal Research Institute, Shenyang, China. A comparison with that from Alfa Co., USA showed no significant differences in results. Electrochemically-etched tungsten wire (0.01” dia. CFW, USA) was used for both STM imaging and nanometer-scale etching. When imaging, the tunneling current was usually controlled at 1 nA with a sample bias voltage of 15~30 mV relative to the tip. To obtain the effective pulsewidth, the “temporary holding” etching procedure was used for surface lithography. The system was run in air and at room temperature.

3. RESULTS AND DISCUSSION

By means of interactive multipoint writing, we wrote the letters “CU” (for Chongqing University) on an HOPG surface, as shown in Fig. 3. The dimension of each letter is about 64nmx64nm, while the letters are formed by a series of pits each about 4 nm in diameter. The asymmetry of the pit shape is partly due to the asymmetry of lateral scan, and partly due to the asymmetry of the atomic structure around the tip apex. We found that the writing often failed at some points, especially if the point followed another selected point within the same scanning line. Otherwise, writing a series of points usually succeeded with high probability. The origin of this phenomenon is not yet clear and is perhaps attributed to some tip change caused by the etching procedure e.g. the tip may adsorb some clusters of atoms or molecules.

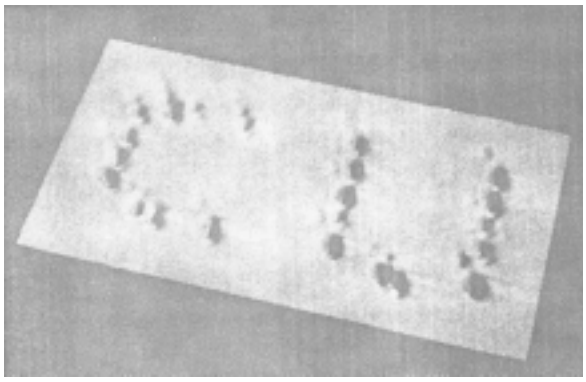

Fig. 3. STM image of a 163 nm x 96 nm HOPG surface after lithography by voltage pulses. The STM imaging conditions are V_b = -30 mV (tip), I_t = 1 nA, scanning speed = 7 Lines/Sec. The voltage pulse for lithography is -5.5 V (tip) with a 0.033 μS width.

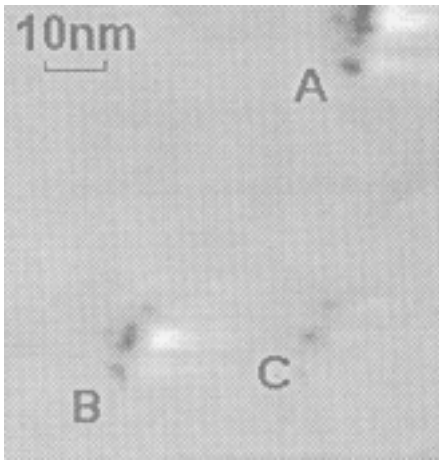

Fig. 4. Influence of pulsewidth on the lithography of an HOPG surface in air. The pulsed voltage amplitude is -5.5 V (tip), and the pulsewidths are 0.030, 0.028, 0.025 μS corresponding to A, B, C points indicated respectively. The STM imaging conditions are V_b = -15 mV (tip), I_t = 1 nA, scanning speed = 7 Lines/Sec.

The voltage threshold for writing pits was obtained as (4.0±0.5) V, consistent with most experiments by other groups. However, in contrast to other experiments, the results shown in Fig. 4 demonstrate that the pit size depends strongly on the pulsewidth. A pulsewidth threshold is also obtained, which varies daily from tip to tip. Considering that our experiments were undertaken in air, we attribute such a variation to the daily change of the air humidity, as is the case of the variation of the voltage threshold [11,13]. The variation of tip condition may be another possible reason. In most cases, a pulsewidth threshold of (0.04±0.01) μS was found, which is far shorter than the pulsewidth used in all HOPG nanolithography to date. This is reasonable given the specially designed experimental setup. A controllable lithography can be achieved only with a pulsewidth near the threshold. Longer voltage pulses usually result in an irregular and widely spread pattern.

One would expect that a pulsewidth below the threshold should result in a much smaller pit. However, experimental results showed that once the pulsewidth was shorter than the threshold, the probability to write pits successfully fell dramatically. One explanation is that the capacitance of the wiring becomes important for shorter pulse, so that the pulse peak amplitude falls below the threshold and the physical changes and/or chemical reactions can not be stimulated. An increase in the applied voltage did improve the writing probability, but also the pit size clearly increased, owing to the increase of the total integrated energy. Another possible explanation is that the pits caused by a voltage pulse shorter than the threshold are too unstable to observe in subsequent STM imaging. The diffusion of various surface atoms may rapidly fill up small pits.

In conclusion, by temporarily holding the tip-sample distance during the etching process, we observed an obvious and short threshold pulsewidth in the nanolithography of HOPG surfaces.

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REFERENCES

[1] J. A. Dagata, W. Tseng, J.Bennett, E.A. Dobisz, J. Schneir and H.H. Harary // *J. Vac. Sci. Technol.* **A10** (1992) 2105.

[2] J. Schneir, J.A. Dagata and H.H. Harary // *J.Vac.Sci.Technol.* **A11** (1993) 754.

[3] Kazuo Terashima, Minoru Kondoh and Toyonobu Yoshida // *J. Vac. Sci.Technol*. **A 8** (1990) 581.

[4] In-Whan Lyo and Phaedon Avouris // *Science* **253** (1991) 173.

[5] R. S. Becker, J. A. Golovchenko and B. S. Swartzentruber // *Nature* **325** (l987) 419.

[6] D. M. Eigler and E. K. Schweizer // *Nature* **344** (1990) 524.
[7] J. S. Foster, J. E. Frommer and P. C. Arnett // *Nature* **33l** (1988) 324.
[8] Phaedon Avouris // *Acc. Chem. Res.* **28** (1995) 95.
[9] K. Matsumoto, M. Ishii, K. Segawa and Y. Oka // *Appl. Phys. Lett.* **68** (1996) 34.
[10] K. Matsumoto // *P. IEEE* **85** (1997) 612.
[11] T. R. Albrecht, M. M. Dovek, M. D. Kirk, C. A. Lang, C. F. Quate and D. P. E. Smith // *Appl. Phys. Lett.* **55** (1989) 1727.
[12] Wataru Mizutani, Junji Inukai and Masatoshi Ono // *Japanese J. Appl. Phys.* **29** (1990) L815.
[13] Reginald M. Penner, Michael J. Heben, Nathan S. Lewis and Calvin F. Quate // *Appl. Phys. Lett.* **58** (1991) 1389.
[14] Philip Moriarty and Greg Hughes // *Appl. Phys. Lett.* **60** (1992) 2338.
[15] S.-T. Yau, D. Saltz, A. Wriekat and M. H. Nayfeh // *J. Appl. Phys.* **69** (1991) 2970.
[16] R. H. Bernhardt, G. C. McGonigal, R. Schneider and D. J. Thomson // *J. Vac. Sci. Technol*. **A 8** (1990) 667.