# Position Control of PbS Quantum Dot by Nanohole on Silicon Substrate

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## Abstract

Hole of nanometer size was processed using the scanning probe microscopy and was successfully applied to the trapping of a PbS colloidal quantum dot. The template method enables position control of a single quantum dot necessary for the manufacturing of quantum information devices.

#### 1. Introduction

Quantum information processing using electrons and photons has attracted much attention as a future technology. Quantum dots (QDs) are regarded as a promising medium for the quantum information devices. The technique to arrange QDs at the desired position is very important from the viewpoint of quantum circuit manufacturing. Using the Stranski-Krastanow-type QDs, many methods to arrange QDs have been investigated. For example, nanoholes on a GaAs substrate were used to grow QDs epitaxially, or the source materials were dropped to grow QDs from a tip of scanning probe microscopy (SPM) onto a substrate, or the pre-processed substrates were found to be effective for the three-dimensional QD arrangement during the epitaxial growth [1-3]. However, these methods have drawbacks such as deformation of QD shape and low quantum efficiency because the methods directly affect the growth process of QDs. On the other hand, some semiconductor QDs can be synthesized in flask. The so-called colloidal QDs are expected to be free from the drawbacks of epitaxial QDs even when they are arranged on a pre-processed substrate. We have reported that SPM lithography was applicable to the preparation of nanoholes of 30 nm in width and 6 nm in depth on a Si substrate [4]. The holes were able to trap colloidal PbS QDs. However, because the diameter of PbS QD was about 6 nm and much smaller than the width of the nanoholes, we could not trap a single QD but trapped a single layer of QDs. In this study, we succeeded to manufacture a nanohole of QD size in both width and depth, and a single QD was trapped by the nanohole.

#### 2. Experimental methods

Figure 1 shows the processing procedure of nanoholes on a Si substrate. Etching masks made of  $SiO_2$  were drawn by the oxidation with SPM. Nanoholes were processed by the reactive ion etching (RIE), and oxide masks were finally removed.  $SF_6$  gas was used for the RIE. Etch selectivity about Si and SiO<sub>2</sub> play an important role to realize high aspect ratio of nanohole. Since edge of the SiO<sub>2</sub> mask is inclined in nanometer order as shown in Fig. 1(b), hole width increases during the RIE. High selectivity is required for deep etching with small expansion of opening. We examined the oxidation condition dependence of the inclination angle of mask edge. We also examined gas flow quantity, process pressure, and RF power during the RIE to improve etch selectivity. PbS QDs were used for the trapping experiments.



Fig. 1 (a) Schematics of nano hole process.(b) Cross section of nanohole during RIE. Etch selectivity is important to prepare nanoholes having the high aspect ratio.

# 3. Results and discussion

The oxidation condition dependence of the edge angle of SiO<sub>2</sub> mask is shown in Fig. 2. High edge angle is preferable to get high aspect ratio of nanoholes. We found that the angle rises by increasing the applied voltage and by decreasing the scanning speed at the SPM oxidation. With the applied voltage of 9.9 V and the scanning speed of 0.25  $\mu$ m/s, we obtained the maximum angle of 2.8°. From the result, we estimated that the selectivity of 20 is necessary to realize the nanohole of QD size with the aspect ratio of 1.

To improve the selectivity, we should restrain the physical effect in comparison with the chemical effect during the RIE [5]. For the purpose, we investigated the effect of RF power, gas pressure, and gas flow rate. Our best selectivity of 12 was obtained with RF power of 40 W, gas pressure of 40 mTorr, and gas flow rate of 30 ccm. With the selectivity, the nanohole equal to QD size was not achievable, and the maximum aspect ratio was expected to

be 0.6. However, even with the aspect ratio, the nanohole having the diameter less than twice that of QD diameter is achievable with the depth equal to QD diameter. Therefore, the selectivity is still enough to manufacture the nanohole available for the single QD trapping.



Fig. 2 Edge angle of  $SiO_2$  masks as a function of (a) the applied voltage and (b) the scanning speed at the SPM oxidation.



Fig. 3 (a) Schematic of center part of the SPM scan pattern to draw small 2 x 2 openings with wide oxide lines.(b) Obtained 2 x 2 nanohole array.

Based on the above findings, we manufactured the nanohole array. Using the scan pattern as shown in Fig. 3(a), the negative mask having 2 x 2 openings was drawn by the SPM. In the drawing of the scan pattern, part of the lines was overwritten because the oxidation line width was 100 nm, i.e., 1/10 of the side of designed openings. Masks completely covered the substrate initially even at the position of nanoholes (see Fig. 1(b)). Figure 3(b) shows 2 x

2 nanohole array after removal of oxide masks. We found that size of the holes varied. The minimum hole has 10 nm x 18 nm opening with the depth of 5 nm.

Using the nanohole, we experimented on the QD trapping. Figure 4 shows SPM images of a nanohole cross-section before and after the QD trapping. It should be noted that the cross-section of nanohole must be triangular because the shape was determined by the mask edge inclination and the etch selectivity (see Fig. 1(b)). We deduced that the actual nanohole cross-section does not have flat bottom and is almost the same as in the SPM images. Taking into account the contours' change at the top of the nanohole in comparison with the shape of QD, we supposed that the hole did not store two QDs, i.e., only one QD was trapped in the nanohole.



(a) before and (b) after QD trapping.

# 4. Conclusions

Aiming at the positioning of a single QD, nanohole of QD size was formed on Si substrate using SPM lithography. By optimizing process conditions to improve the cross-sectional shape of SPM oxidation lines and Si/SiO<sub>2</sub> selectivity in the RIE, we realized the fine negative-type SiO<sub>2</sub> masks suitable for the processing of nanoholes for the single QD trapping. SPM images suggested the successful trapping of a single QD with the processed nanohole.

# References

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