

# Bottom-up approach to silicon nanoelectronics

Hiroshi Mizuta<sup>a,b,\*</sup>, Shunri Oda<sup>a,b</sup>

<sup>a</sup>*Quantum Nanoelectronics Research Center and Department of Physical Electronics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8552, Japan*

<sup>b</sup>*SORST JST (Japan Science and Technology), Tokyo, Japan*

Received 24 February 2006; accepted 10 October 2006

Available online 15 February 2007

## Abstract

This paper presents a brief review of our recent work investigating a novel bottom-up approach to realize silicon-based nanoelectronics. We discuss fabrication technique, electronic properties, and device applications of silicon nanodots as a building block for various nanoscale silicon devices.

© 2006 Published by Elsevier Ltd.

**Keywords:** Silicon; Quantum dot; Nanoelectronics

## 1. Introduction

Over the past few decades, the performance of silicon-based VLSI circuits has been steadily improved by scaling down device dimensions, and a nearly exponential growth of microelectronics capabilities has been achieved. However, maintaining this top-down miniaturization trend is becoming exceedingly hard due to fundamental physical and technological limitations as well as the economical limitation, although the International Technology Roadmap of Semiconductors (ITRS) now predicts that the physical gate length of high-performance MOSFETs will reach sub-10-nm in 2016. By contrast, the use of organic molecules as a building block for nanoscale devices has attracted much attention because the precisely controlled nanostructures may be formed cheaply by utilizing self-assembly of molecules. This bottom-up technology can potentially overcome the inherent problem of the present silicon top-down technology. The conductivity of the organic molecular structures is, however, still much lower than that of silicon as the electron transport along the single molecule is basically governed by hopping conduction.

Silicon nanodots (SiNDs) and nanowires (SiNWs) [1–4] may provide a solution to these issues by meeting the requirements of both bottom-up organization and superior carrier transport properties. As those silicon nanostructures can be formed on non-Si substrates, such as glass and plastic, the Si-based bottom-up approach may lead to high-performance and large-area electronics. In addition, the zero- and one-dimensional nature of electronic states in the individual SiNDs and SiNWs realizes new electronic and photonic properties, which are not achieved with bulk silicon. Combining the bottom-up approach with the conventional top-down Si technologies enables us to explore silicon nano-, micro-, and macroelectronics on a common technical footing.

In this paper, we focus on the SiNDs in particular and present our recent studies on fabrication technique, unique electronic properties, and novel device applications.

## 2. Bottom-up silicon nanostructure fabrication

### 2.1. Formation of nanocrystalline silicon dots

For fabricating SiNDs, we have studied three different techniques. The first method is to use a very thin nanocrystalline (nc) Si film with the size of the grains down to a few nanometers. The nc-Si films can be formed either from

\*Corresponding author. Tel.: +81 3 5734 3591.

E-mail address: [mizuta@pe.titech.ac.jp](mailto:mizuta@pe.titech.ac.jp) (H. Mizuta).

an amorphous Si film with solid-phase crystallization (SPC) or by using a very high frequency (VHF) plasma-enhanced chemical vapor deposition (PECVD) at a low temperature [5]. In the SPC films, the individual grains are usually columnar shaped, and the grain boundaries (GBs) between adjacent grains contain carrier trap states due to dangling bonds. By contrast, in the PECVD films, the individual grains are more spherical, and the GBs are formed by a-Si:H layers between grains. The second approach is to use porous Si [6] formed by using photoanodization of the Si substrate. The surface of the SiND islands formed in the substrate can be oxidized selectively by electrochemical oxidation. The formation of a linear chain of SiND islands with a diameter as small as 5 nm has been observed [7].

The most recent approach is a VHF plasma-enhanced deposition of silane ( $\text{SiH}_4$ ) with a hydrogen gas pulse sequence [8]. This technique facilitates separating the nucleation and crystal growth process and enables fabrication of single crystalline SiNDs (Fig. 1) with diameters less than 10 nm. The nucleated SiNDs are grown in the  $\text{SiH}_4$  plasma in the intervals between  $\text{H}_2$  gas pulses, and dot diameter is therefore determined by the growth time of the SiNDs in the plasma cell. Using this method, we fabricated SiNDs with diameters of around 8 nm and a very narrow size spread ( $\pm 1$  nm). Using a self-limiting oxidation process, it is possible to reduce the dot size further down to 4 nm. The interparticle tunnel barriers can also be formed by *in situ* oxidation or nitridation in a controlled manner.

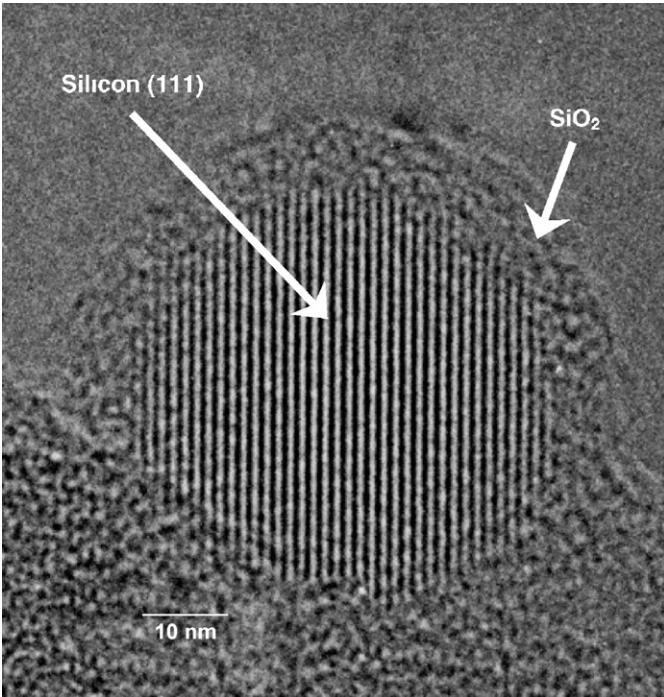


Fig. 1. A TEM image of a single crystalline SiND covered with a thin  $\text{SiO}_2$  layer.

## 2.2. Combining bottom-up and top-down approaches for fabricating nanoscale device structures

Integrating the fabricated SiNDs, either over a large area or in a local area, is a very challenging issue. Various techniques are currently under investigation, for example, the dispersion solution drop and evaporation method [9], the Langmuir–Blodgett (LB) method, and the nanotemplate method. We also examined a novel method of fabricating nanoscale devices by conducting the self-assembly of the SiNDs on patterned nanoelectrodes [10,11]. For preparing the SiND dispersion solution, substrates with deposited SiNDs were soaked into solvents such as methanol or isopropanol, and the ultrasonic treatment was applied for a few tens of seconds. The SiND solution was then condensed by evaporating the solvent a fraction.

The nanoelectrodes were fabricated by using electron beam lithography on the heavily doped n-type silicon on insulator (SOI) substrate with a thickness of about 50 nm and a buried oxide (BOX) thickness of 200 nm. This structure gives a good areal contrast of hydrophobic (Si) and hydrophilic ( $\text{SiO}_2$ ) surfaces and works as a template for the following SiND assembly process. We used the drop and evaporation technique [9] to assemble the SiNDs from the dispersion solution by using the lateral capillary meniscus force, which operates at the point where the three phases of the liquid, air, and SiND meet. Fig. 2 shows the SiNDs assembled on the SOI substrate with a patterned nanogap of about 35 nm. We observed that SiNDs remained only in the  $\text{SiO}_2$  regions and were assembled near the nanogap, resulting in a SiND channel between the electrodes. This trend was observed in common over a large area. This method may be useful to fabricate the SiND-based nanoscale transistors, and we believe that it is possible to form a channel with few SiNDs or even a single dot by reducing the density of SiNDs and optimizing evaporation conditions.

## 3. Electron transport properties of Si nanodots

### 3.1. Resonant tunneling via single Si nanodot

Strong quantum confinement effect in a SiND is the key to realize Si-based resonant tunneling devices. We

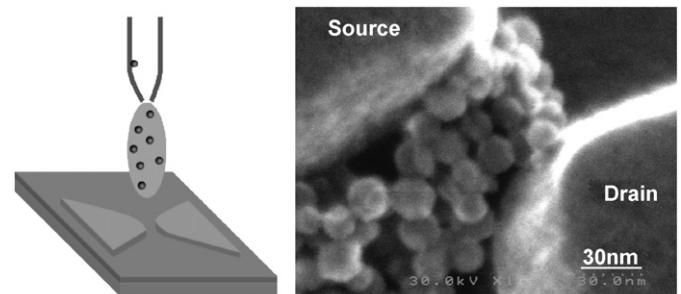


Fig. 2. SiNDs assembled on the nanoelectrodes by using the dispersion solution method.

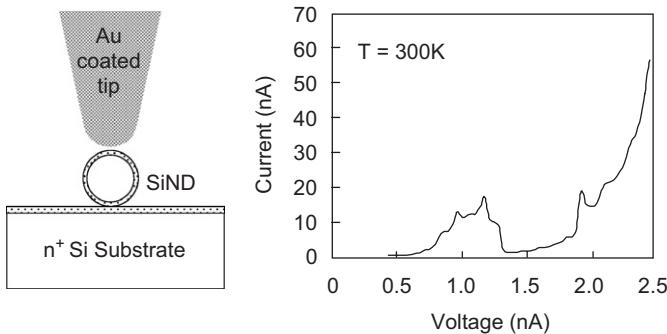


Fig. 3. Tunneling current via a single SiND measured at room temperature by using contact mode AFM.

characterized tunneling current through a single SiND by using the contact mode AFM [12]. The SiNDs were sparsely distributed on the n-type [100] Si substrate with a surface density of  $1.4 \times 10^8 \text{ cm}^{-2}$ . For the cantilever, we used a silicon tip coated with gold. We performed contact AFM scanning with  $1 \times 1 \mu\text{m}^2$  scan area to generate the topography image. The AFM measurement was performed by selecting a single SiND from the topographical view.

*I*–*V* characteristics were measured for a single SiND at room temperature [11]. Fig. 3 shows the typical *I*–*V* curve observed when the sample was positively biased from 0 to 4.5 V. Negative differential conductance (NDC) was clearly observed at a bias voltage of about 1 V, and the peak-to-valley (*P*/*V*) current ratio was as large as about 17. As SiND-based resonant tunneling structures can be integrated with conventional MOSFETs, the observed multiple NDC characteristics may be used for making Si-based novel functional devices such as tunnel-based SRAM and resonant tunneling transistors.

### 3.2. Coulomb blockade and electron interaction in coupled double Si nanodots

Charge quantization and Coulomb blockade have also been studied intensively for SiNDs. Coulomb oscillation of tunneling current has been observed at room temperature by using point-contact single-electron transistors (PC-SETs) with very few SiNDs in the channel. Electrostatic and coherent coupling effects have also been studied recently for strongly coupled double SiNDs by using PC-SETs fabricated on an nc-Si thin film. The PC-SETs with a very small channel, with  $30 \times 30 \text{ nm}^2$  in lateral dimensions, were formed on a 40-nm thick nc-Si film with a lateral grain size of 20–25 nm. The electrostatic potential on the grains is controlled via the bias applied to two side gates.

The PC-SETs exhibited delocalization of the electron wavefunctions over the coupled double dots via a very thin tunnel barrier. A plot of the device conductance at 4.2 K as a function of the two side gate voltages shows single-electron conductance peaks, which partially form an electron stability diagram for two charging dots (Fig. 4).

The peak lines in this plot (white dotted lines) show strong splitting (a dotted circle) caused by electrostatic

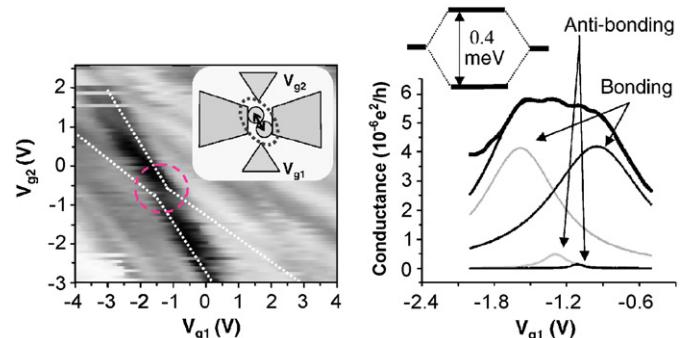


Fig. 4. Coherent electron coupling observed for the tunneling current via strongly coupled double SiNDs.

interactions when the energy levels in the two dots are in resonance [13]. In this strong coupling region, we observed that the characteristics are decomposed into four Lorentzian peaks: two main peaks with two small peaks (Fig. 4; right figure) [14]. This is attributed to the tunnel coupling across two adjacent double dots, resulting in bonding- and anti-bonding-like resonance peaks. Tunnel splitting obtained from the peak separation is about 0.4 meV, which is from a few times to an order-of-magnitude larger than the value reported previously in GaAs/AlGaAs quantum dots. Such quasi-molecular states may be used to realize a Si-based charge qubit.

### 3.3. Phononic band formation in Si nanodot array

Silicon and SiO<sub>2</sub>, the key players in the present VLSIs, now combine in a different way to offer new functional applications in electronics and mechanics. Electron transport properties of the SiNDs interconnected with thin oxide layers have recently attracted growing attention due to the experimental observation of ballistic electron emission [15] as well as the theoretical study of phonon depletion [16]. The electronic and phononic states have been recently investigated for a one-dimensional array of SiNDs interconnected with thin oxide layers as shown in Fig. 5. This acoustic heterostructure has a wide variety of interesting features such as phonon bandgap and phonon confinement. Changing the thickness of the oxides controls the energy range of the bandgap. It is also interesting to note that the energy dispersion for high-energy phonons is flat. Such phonons are confined in SiNDs.

Another interesting feature of this structure is the reduction of the electron–phonon scattering potential, which is written as  $H_{\text{el-ph}}(x) = D_{\text{aco}} \partial S(x) / \partial x$  where  $S(x)$  and  $D_{\text{aco}}$  denote phonon wave function and the coupling constant, respectively. The first derivative of  $S(x)$  is also known as the strain. Fig. 6 compares the strain in the acoustic heterostructure (circles with solid line) and conventional Si (broken line). Note that the oxide layers ‘absorb’ the strain from the SiNDs [17]. This is reasonable because the oxide layers are ‘softer’ than SiNDs (the Young’s modulus of Si and oxide are 180 and 70 GPa,

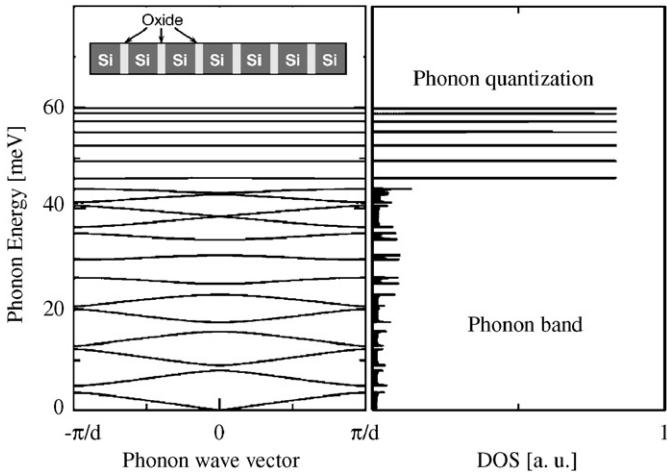


Fig. 5. A concept of SiND/SiO<sub>2</sub> acoustic heterostructure and calculated phonon energy dispersion (left) and associated phonon density of states (right).

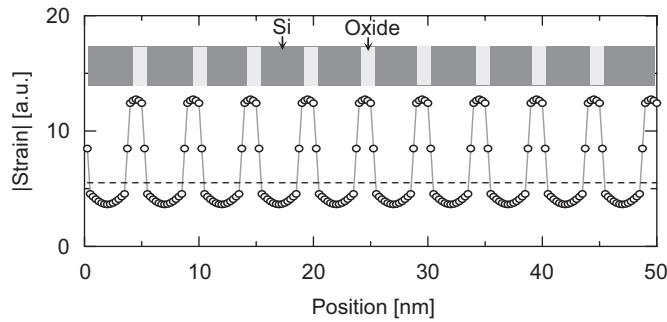


Fig. 6. Phonon accumulation in SiO<sub>2</sub> and associated phonon depletion in SiNDs obtained for acoustic phonon modes.

respectively). As the coupling constant  $D_{aco}$  in the oxide is smaller than that in Si, the strain absorption effect reduces the scattering potential over the entire region compared with that of Si. It was shown theoretically [18,19] that the electron energy loss rate is suppressed significantly in the vicinity of the miniband bottom energy.

#### 4. Novel functional device applications based on silicon nanodots

Based on the new properties shown in Section 3, we have pursued various device applications such as ballistic electron emission devices, light-emitting devices, single-electron transistors [20], SiND memory with a long data retention time [21], and a nonvolatile nanoelectromechanical system (NEMS) memory. Two unique device applications are introduced in the following sections.

##### 4.1. Ballistic electron emission device

As one of the promising applications of the SiNDs, we investigated the SiND-based electron surface emitter [22]. Unique phonon properties of the array of SiNDs covered

with SiO<sub>2</sub>, shown in Section 3.3, the lead to ballistic electron emission phenomenon. An electron emitter device was fabricated by using a 150-nm-thick layer of Si nanodots deposited on n<sup>+</sup>-Si substrate and a very thin Au top electrode (Fig. 7). When the voltage is applied across the SiND layer, only electrons with energy higher than the Au work function  $W_{Au}$  of ~5 eV are emitted into vacuum through the top electrode. Both diode and emission currents were measured as a function of the extraction voltage  $V_{ex}$ , and the emission efficiency could be as large as 5%.

We also investigated the energy distribution of emitted electron by using a conventional ac-retarding-field analyzer. As shown in Fig. 8, we found that the energy distribution of the emitted electrons is non-Maxwellian in contrast with those observed for conventional cold emitters. Maximum electron energy agrees approximately with  $eV_{ex} - W_{Au}$ , and the peak energy varies in proportion to  $V_{ex}$ . These results show that fractional electrons travel throughout the SiND layer in a quasi-ballistic manner and emit into vacuum.

#### 4.2. Nanoelectromechanical memory device

A new nonvolatile memory concept has recently been proposed, based on the bistable operation of the sub-μm-long

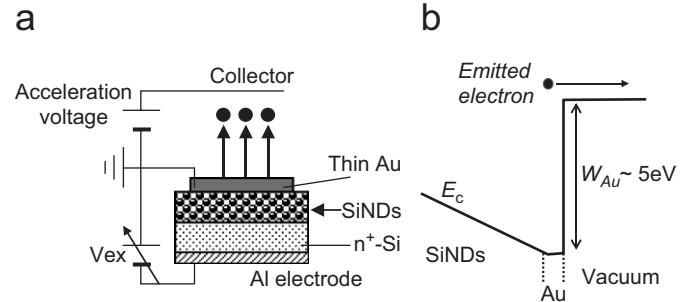


Fig. 7. (a) Schematic ballistic electron emitter based on stacked SiNDs and (b) the energy-band diagram near the surface of the SiND layer.

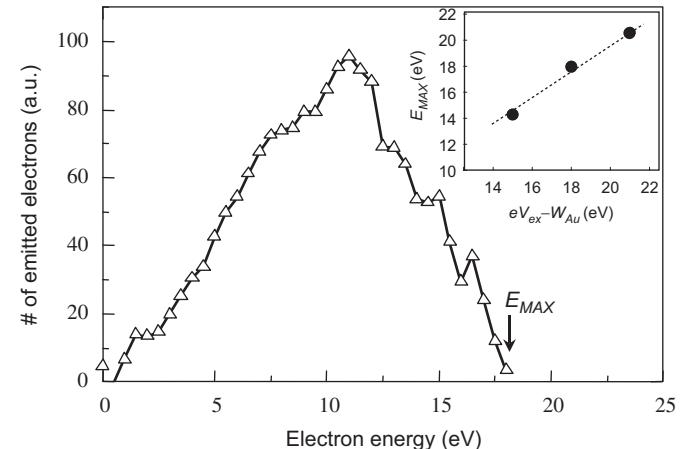


Fig. 8. Energy distribution of electrons emitted from the SiND layer measured at the external bias voltage  $V_{ex}$  of 23 V. The inset plots the maximum electron energy as a function of  $eV_{ex} - W_{Au}$ .

NEMS structure, combined with the SiNDs (Fig. 9; above) [23]. It features a suspended  $\text{SiO}_2$  beam, which incorporates the SiNDs as single-electron storage (Fig. 9; below). The beam may be moved via the gate electric field, and its positional displacement is sensed via a change in the drain current of the MOSFET underneath.

A free-standing  $\text{SiO}_2$  single beam was first fabricated using a Si undercut etching technique. Most fabricated samples show the beams bent upwards as a result of

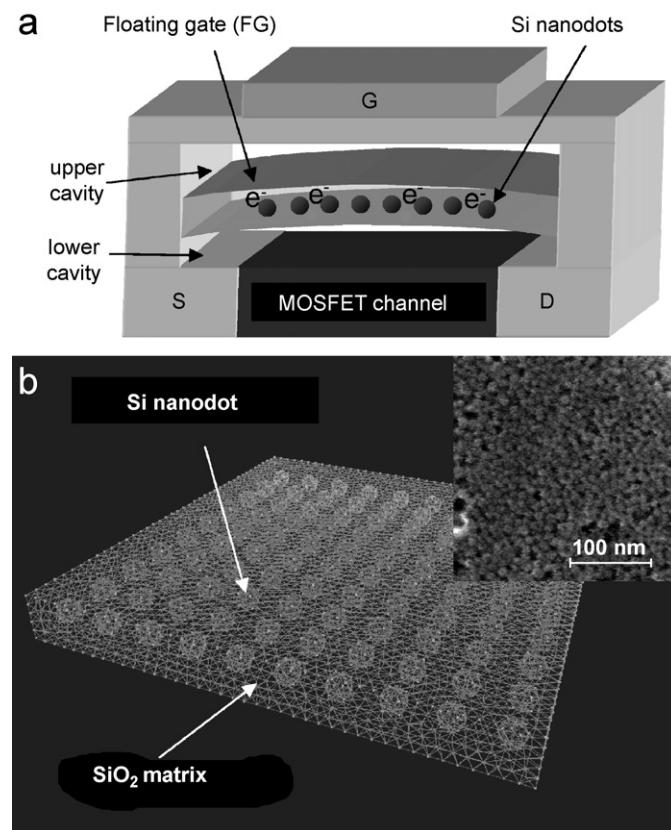


Fig. 9. (a) A schematic illustration of nanoelectromechanical memory structure and (b) a floating gate incorporating SiND array.

mechanical stress, stored in the  $\text{SiO}_2$  being released. The mechanical bistability of the beam was demonstrated by using the nanoindentor-type loading system. Also, a top gate electrode has been fabricated successfully above the  $\text{SiO}_2$  (Fig. 10).

The switching speed and power of the beam have been studied intensively by using a 3D FEM simulation [24]. The switching speed increases approximately in inverse proportion to the beam length  $L$  and goes beyond 1 GHz in the sub- $\mu\text{m}$  regime. The switching voltage of less than 15 V has been demonstrated recently for an optimized beam structure along with the scaling properties of the beam. In contrast with other emerging nonvolatile memories such as MRAM or PCRAM, the NEMS memory can still be manufactured within the conventional Si technologies. With further optimization, it may exhibit excellent characteristics comparable to them.

## 5. Conclusion

We have studied SiNDs as a promising building block for Si nanoelectronics. SiNDs with diameters as small as 5 nm were fabricated by using VHF plasma CVD to achieve an interface tunnel layer thickness of 1 nm. In addition, the SiND assembly technique was successfully combined with the nanofabrication technique to build nanoscale transistors with SiNDs as a channel. A variety of new functions were found for SiNDs: ballistic electron emission, single-electron charging effects, and quantum mechanical coupling between two adjacent dots, which have never been observed before in bulk silicon. Based on these unique material properties, we have explored various novel device applications such as a ballistic surface electron emitter and a high-speed and nonvolatile nanoelectromechanical memory. An innovative fusion of top-down and bottom-up silicon technologies may lead to a high-performance and low-cost material platform for macro-, micro-, and nanoelectronics.

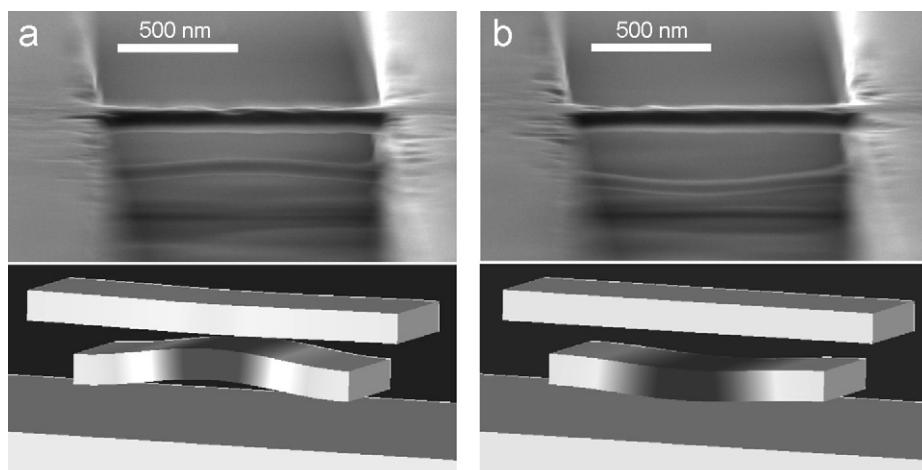


Fig. 10. Bistable (a) upward-bent and (b) downward-bent states NEMS FG: fabricated (top) and simulated (bottom).

## Acknowledgments

The authors are very grateful to Dr. Y. Tsuchiya, Dr. K. Usami, Dr. M. Khalafalla, Dr. S. Huang, Dr. K. Nishiguchi (now of NTT Basic Res. Lab.), Mr. K. Takai (now of Toshiba Corp.), Mr. T. Nakatsukasa (now of Toppan Printing Co. Ltd.), Mr. A. Surawijaya, Mr. N. Momo, Mr. T. Nagami, Mr. A. Tanaka, Mr. G. Yamahata, Mr. J. Ogi of Tokyo Institute of Technology, Dr. Z.A.K. Durrani of Cambridge University, Dr. S. Uno of Nagoya University and Prof. N. Mori of Osaka University for their valuable technical contributions, and Dr. S. Saito, Dr. T. Arai of Central Research Laboratory, Hitachi Ltd., Dr. T. Shimada of Quantum 14 Co. Ltd., Prof. K. Nakazato of Nagoya University, Prof. N. Koshida of Tokyo University of A&T and Dr. T.A. Armour of Nottingham University for very useful discussions.

## References

- [1] Y. Cui, C.M. Lieber, *Science* 291 (2001) 851.
- [2] Y. Huang, X. Duan, Q. Wei, C.M. Lieber, *Science* 291 (2001) 630.
- [3] X. Duan, C. Niu, V. Sahi, J. Chen, J.W. Parce, S. Empedocles, J.L. Goldman, *Nature* 425 (2003) 274.
- [4] S. Piscanec, M. Cantoro, A.C. Ferrari, J.A. Zapien, Y. Lifshitz, S.T. Lee, S. Hofmann, J. Robertson, *Phys. Rev. B* 68 (2003) 241312.
- [5] T. Kamiya, K. Nakahata, Y.T. Tan, Z.A.K. Durrani, I. Shimizu, *J. Appl. Phys.* 89 (2001) 6265.
- [6] N. Koshida, H. Koyama, *Appl. Phys. Lett.* 60 (1992) 347.
- [7] A.G. Cullis, L.T. Canham, P.D.J. Calcott, *J. Appl. Phys.* 82 (1997) 909.
- [8] S. Oda, M. Otabe, *Mater. Res. Soc. Proc.* 358 (1995) 721.
- [9] Y. Tsuchiya, T. Iwasa, A. Tanaka, K. Usami, H. Mizuta, S. Oda, MRS Spring Meeting, April 2004.
- [10] A. Tanaka, G. Yamahata, Y. Tsuchiya, K. Usami, H. Mizuta, S. Oda, *Curr. Appl. Phys.* (2005) in press.
- [11] A. Tanaka, G. Yamahata, Y. Tsuchiya, K. Usami, H. Mizuta, S. Oda, 5th IEEE Conference on Nanotechnology, 2005.
- [12] A. Surawijaya, H. Mizuta, S. Oda, Extended Abstract of 2005 International Conference on Solid State Devices and Materials, 2005, p. 180.
- [13] M. Khalafallah, Z.A.K. Durrani, H. Mizuta, *IEEE Trans. Nanotechnol.* 2 (2003).
- [14] M. Khalafallah, H. Mizuta, Z.A.K. Durrani, *Appl. Phys. Lett.* 85 (2004) 2262.
- [15] N. Koshida, X. Sheng, T. Komoda, *Appl. Surf. Sci.* 146 (1999) 371.
- [16] E.P. Pokatilov, D.L. Nika, A.A. Balandin, *Appl. Phys. Lett.* 85 (2004) 825.
- [17] S. Uno, K. Nakazato, S. Yamaguchi, A. Kojima, N. Kojima, H. Mizuta, *IEEE Trans. Nanotechnol.* 2 (2003) 301.
- [18] S. Uno, N. Mori, K. Nakazato, N. Koshida, H. Mizuta, *J. Appl. Phys.* 97 (2005) 113506.
- [19] S. Uno, N. Mori, K. Nakazato, N. Koshida, H. Mizuta, *Phys. Rev. B* 72 (2005) 035337.
- [20] A. Dutta, S. Oda, Y. Fu, M. Willandar, *Jpn. J. Appl. Phys.* 39 (2000) 4647.
- [21] S. Huang, K. Arai, K. Usami, S. Oda, *IEEE Trans. Nanotechnol.* 3 (2004) 210.
- [22] K. Nishiguchi, X. Zhao, S. Oda, *J. Appl. Phys.* 92 (2002) 2748.
- [23] Y. Tsuchiya, K. Takai, N. Momo, S. Yamaguchi, T. Shimada, S. Koyama, K. Takashima, Y. Higo, H. Mizuta, S. Oda, 2004 Silicon Nanoelectronics Workshop, 2004, p. 101.
- [24] T. Nagami, N. Momo, Y. Tsuchiya, S. Saito, T. Arai, T. Shimada, H. Mizuta, S. Oda, 2005 Silicon Nanoelectronics Workshop, 2005, p. 94.