SURFACE-TEMPERATURE CONTROL OF SILICON NANOWIRES IN DRY AND LIQUID CONDITIONS

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ABSTRACT

In this paper we present the results of surface temperature control of silicon nanowires by using fluorescent thermometry at the nanometer scale. Rhodamine B is one of the stable fluorescent molecules, which rely on the characteristic of temperature-dependent change in fluorescent intensity, and it was used for nano-scale surface temperature sensing interface. The resistive heating on Si nanowires was carried out with applying voltage potential of 6 ~ 12 V. Surface-temperature measurement was performed by converting the changes in fluorescent intensity with calibration curve of Rhodamine B. The temperature at the central line along nanowires increasing from 30 degrees to 35~70 degrees was observed.

INTRODUCTION

Silicon nanowires have attracted much attention for many years because they are most attractive from the point of view of high sensitivity and surface-modifying for various sensors. Recently, silicon nanowires are widely applied for biosensors [1]. But a few attempts [2] were made so far to control their temperature although this parameter has been shown to be crucial when dealing with biochemical reactions [3]. There are several methods which measure temperature under the micrometer scale, for example Raman spectroscopy and Infrared thermometry. But considering the spatial resolution and cost of setup, fluorescent thermometry has considerable promise as thermometer at the sub-micro scale. It relies on the measurement of the photoluminescent light emitted by a probe in order to determine the temperature. Since the probe used in fluorescent thermometry transmit information via light, the method is useful as a non-contact approach in the characterization of temperature.

Using a fluorescent probe, Rhodamine B, which shows fluorescent intensity dependence as a function of the temperature [4], Löw *et al.* demonstrated the possibility to monitor the temperature of nickel nanowires by fluorescence thermometry in dry condition [5]. However, this work showed several issues, such as a high thermal inertia. Besides, nickel is difficult to functionalize and its reflectivity makes difficult the microscopic studies of low fluorescence signals.

From these backgrounds, we think that it is important to control surface-temperature of silicon nanowires in dry as well as in liquid conditions. The main advantage of silicon nanowire heating device is the capability for fast temperature cycling due to the lower thermal mass and heat capacitance.

2. MATERIALS AND METHODS

Fabrication of the nanowires

We fabricated the silicon nanowires from Silicon-on-Insulator (SOI) wafer with conventional microfabrication processes only [6]: photolithography, anisotropic wet etching, local thermal oxidation of silicon (LOCOS), and removal of silicon dioxide. The basic principles and processes are shown in figure 2. First, wafer cleaning and silicon nitride deposition were carried out (fig. 2a and 2b). Approximately 100 nm-thick silicon nitride (Si_3N_4) film was deposited on the surface of SOI substrate by using Low-Pressure Chemical Vapor Deposition (LPCVD). This silicon nitride layer was used as a masking layer in the silicon anisotropic KOH solution etching (Fig. 2c) and local oxidation of silicon process (Fig. 2d). Next, in order to remove silicon nitride film on the pre-wire structure, photolithography and Reactive Ion Etching (RIE) process were carried out (Fig. 2e and 2f). After that, anisotropic KOH solution etching was performed again to remove silicon partially in the pre-wire structure (Fig. 2g). Finally remaining Si_3N_4 film was removed by RIE process (Fig. 2h). As additional process, sputtering process was carried out to change the surface characteristic. The surface of silicon nanowires was changed from silicon to silicon dioxide to attach the biological molecules.

The most important point of this process is that the width and thickness of fabricated silicon nanowires would be determined by the thickness of top silicon layer of SOI substrates only. The final length, width and thickness of the nanowires are 20 μm, 360 nm and 240 nm, respectively. Due to the anisotropic etching process dependant on Si crystalline, the shape of cross sectional nanowire is a triangle. The SEM images of silicon nanowires are shown in figure 1.

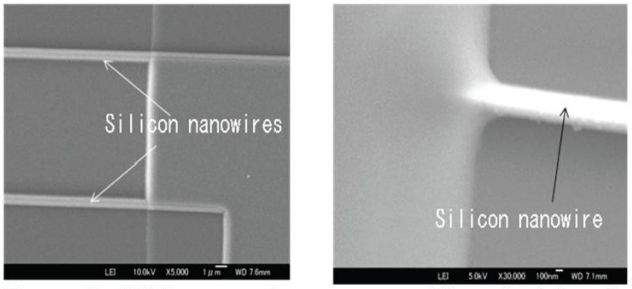

Figure 1: SEM images the nanowires. Their final length, width and thickness are 20 μm, 360 nm and 240 nm, respectively.

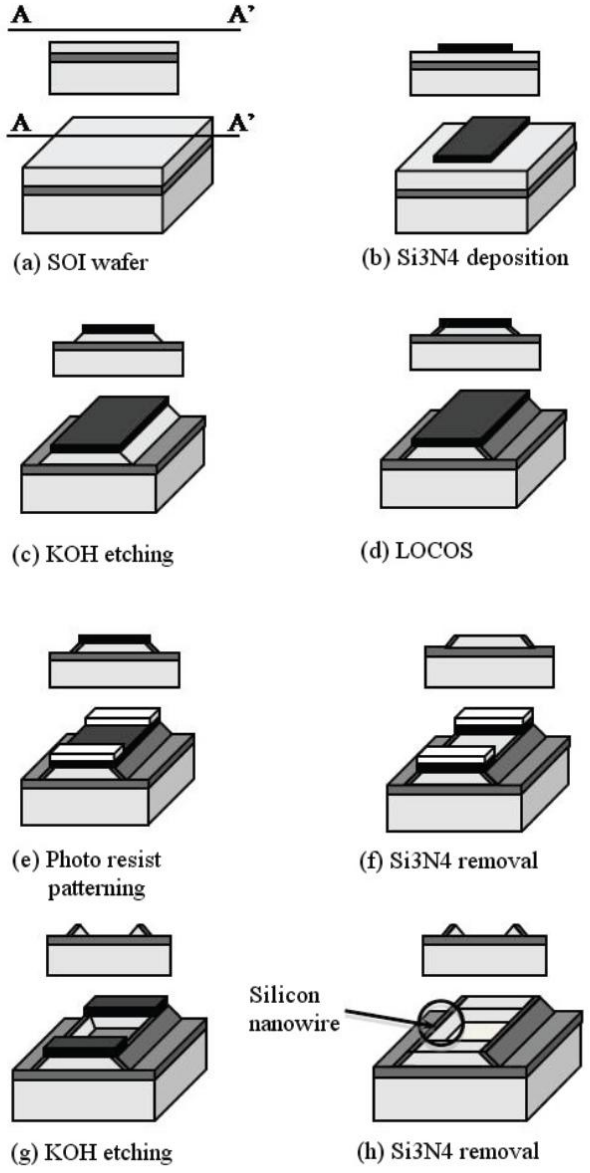

Figure 2: Fabrication process of the nanowires: (a) Preparation of the SOI substrates; (b) Silicon nitride deposition; (c) Top silicon layer etching by KOH; (d) Top silicon layer oxidation (LOCOS); (e) Lithography; (f) Si_3N_4 film removal; (g) Nanowire formation by KOH etching; (h) Removal of remaining silicon nitride.

Finally, we decrease the resistivity of the nanowires by a p-type ion implantation of silicon (ULVAC UP-150, Japan, 3×10^{15} ion/cm^2, 15 μA) in order to improve the performances of the nanowires toward resistive heating.

Functionnalization of the Nanowires by Rhodamin-B

For the experiment in air (dry condition), the Rhodamine B molecules (Molecular Probes, Invitrogen) are dissolved in deionized water (Millipore) and simply dried on the surface of the nanowires before carrying out the measurements.

However, in order to perform the measurement with the nanowires immersed in an aqueous buffer (*liquid condition*), rhodamine has to be firmly attached to surface to avoid its spontaneous desorption enhanced by the high temperature [7]. Instead of using bare rhodamine, we use a functionnalization route involving the biotin-streptavidin complex since it is easy to handle [8] and it has been shown to be thermally stable until a temperature of 95 degrees, which is sufficient for our application [9].

We first adsorb a positive polyelectrolyte (poly-L-lysine, *Sigma-Aldrich*) to which we attach a biotin derivative Biotin-$(C_2H_4O)_3$-NH_2 ($C_{16}H_{30}N_4SO_4$, Pierce) and finally adsorb a protein, rhodamin-streptavidin (Molecular Probes, Invitrogen), thanks to its high chemical affinity with biotin [10].

Fluorescent Thermometry Setup

Fluorescent measurements were made using a BX-51 upright fluorescence microscope and a conventional mercury lamp from Olympus, Japan. In order to capture the fluorescence for digital analysis, we use an EM-CCD (electron-multiplying CCD) camera (Cascade II 512, Photometrics, USA) connected to a computer. During the experiments we use a thermal plate (MATS-1002RO, TOKAI HIP Co., Ltd., Japan) to set the temperature of the silicon support of the nanowires to 30 degrees as a reference. The schematic diagram of fluorescence thermometry is shown in figure 3.

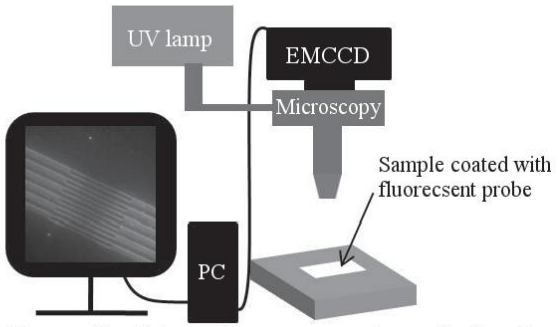

Figure 3: Schematic representation of the fluorescent thermometry setup under the microscope we use to perform the temperature mapping of the nanowires.

The Al-electrodes were patterned on the substrate by evaporation and lithography. Next, the silicon nanowire tip was connected to a commercial PCB board by using a wire bonder (MB-2100, Nippon Avionics Co., Ltd., Japan). The resistive heating on nanowires was carried out with applying voltage potential by a DC power supply (R6240A DC Voltage current source/monitor, Advantest, Japan). In dry condition, silicon nanowire tip was covered by a coverslip, which fixed with tapes. On the other hand in liquid condition, the silicon nanowire tip was sealed with commercial nail lacquer after this process to avoid evaporation of the aqueous buffer.

3. RESULTS

Calibration of the Fluorescence Intensity Measurement

To calibrate the measurement, we measure the fluorescence intensity of a functionalized and oxidized silicon surface in *dry* and *liquid* condition when varying its temperature with an accurate hot-plate mounted on the microscope. Since the intensity of fluorescence is decreasing with the increasing temperature, we calculate the ratio

$$r = \frac{I_T}{I_{30}}$$

of the intensity I_{30} at 30 degrees to the intensity I_T at higher temperatures. On figure 4, we plot the intensity ratios as a function of the hot-plate temperature. In air and in liquid, the intensity decrease is similar and scales linearly with the temperature. In the following experiments we use these data to calibrate the temperature of the nanowires.

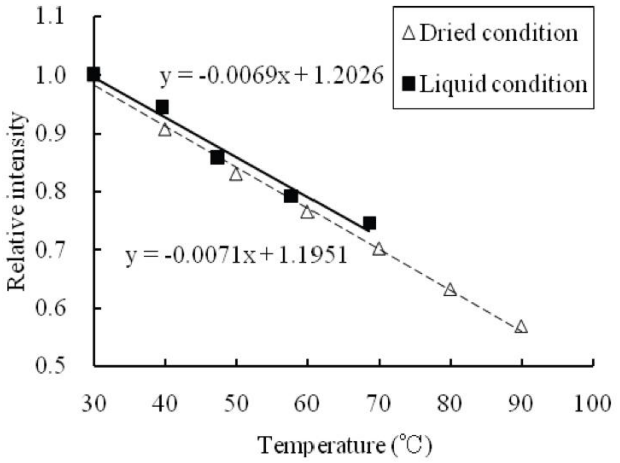

Figure 4: Calibration curve of the fluorescent intensity of Rhodamine B molecules as a function of temperature in dry (△) and liquid (■) conditions. In both cases, the relative intensity scales linearly with the temperature.

Temperature Mapping of the Nanowires

In dry condition, we measured the intensity of fluorescence of silicon nanowires coated with Rhodamine B when a voltage ranging from 0 to 13 V is applied (Fig. 5). We can see silicon nanowires clearly with no voltage in the left image. But in the right image, because of applied voltage, surface temperature of silicon nanowires was increased. As a result the white parts of silicon nanowires changed black.

From the calibration data, we are able to make the temperature mapping of the silicon nanowires when the tension is increased. First, the intensity of background was subtracted from the averaged image. Next, the intensity of every pixel on the applied voltage images was divided by the no voltage images to discover the change of intensity, which means the ratio of intensity. At last, the changes in intensity were converted to changes in temperature by utilizing calibration curve of fluorescent intensity of Rhodamine B.

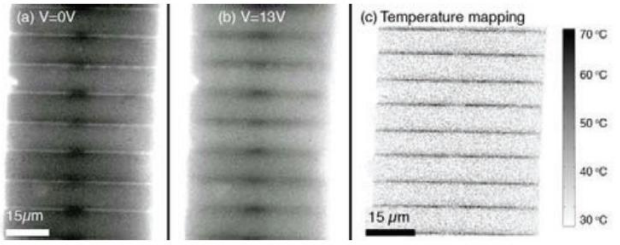

Figure 5: Fluorescence microscopy images of silicon nanowires (Length=50 μm) coated with Rhodamine B molecules in dry condition with no voltage (a) and 13 V (b). The fluorescence intensity decreases with the increase of temperature.

The distribution of surface temperature along and cross the silicon nanowires is shown on figure 6. The temperature of the wires ranges from 35 to almost 70°C.

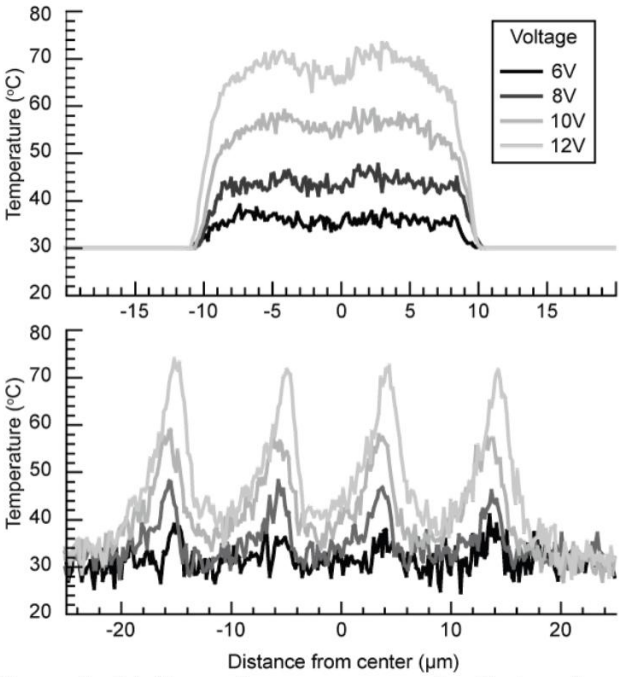

Figure 6: (a) The surface temperature distribution along single silicon nanowires as a function of the applied voltage. The temperature is almost constant over the full length of the nanowire and ranges between 35 and 70 degrees. (b) Comparison of the temperature distribution over four adjacent nanowires on the same silicon substrate as a function of the applied voltage. The nanowires have the same temperature.

On figure 7, Mean temperature of the nanowires plotted as a function of the electrical tension applied. The temperature increases linearly over the whole tension range. Based on these results, it is clear to understand that the surface temperature of silicon nanowires increased corresponding to the increased applied voltage potential.

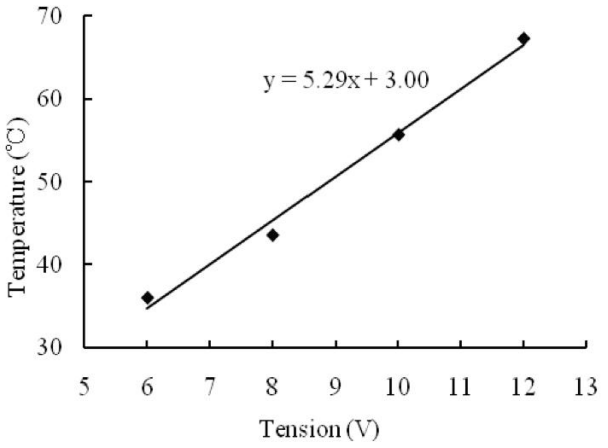

Figure 7: Mean temperature of a nanowire as a function of the electrical tension. The temperature increases linearly over the whole tension range.

4. CONCLUSION

In this article, we measure the temperature of silicon nanowires as a function of the applied voltage by fluorescence thermometry. After a calibration step of the measurement in *dry* and *liquid* condition, we are able to make the spatial mapping of the temperature of the nanowires, and we show that this temperature ranges linearly until 70 degrees with the applied voltage in *dry* condition. The next step will be to perform the same measurement in *liquid* condition in order to use the devices for biomolecular studies.

5. ACKNOWLEDGMENT

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6. REFERENCES

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