Growth of silicon self-assembled nanowires by using gold-enhanced CVD technology


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Abstract. In spite of the fact that, a great deal of experimental research has been published, there is a lack of good understanding of silicon nanowires growth to exert control over important properties of the system, though it presents the simplest system like gold on silicon substrate. In the current research, to find the best conditions to grow silicon nanowires with presupposed properties, we studied various technological regimes both of growth-seed formation and conditions of silicon nanowires growth.

Keywords: silicon nanowires, gold enhanced growth, CVD-technology.

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1. Introduction

Silicon nanowires (SiNWs) are promising material for applications in micro-, opto- and bio-electronics and information technologies. In the recent decade, a special attention is particularly paid to bio- and medical applications. The authors of [1] demonstrate efficient delivery of biomolecules into mammalian cells using surface-modified vertical silicon nanowires. The method relies on the ability of silicon nanowires to penetrate through the cell membrane and subsequently release surface-bound molecules directly into the cell cytosol. SiNWs based resistors used as bacteria sensors [2]. These resistors are promising to monitor contamination when controlling the environment for hygiene and can be fabricated in a simple and low-cost technological ways. Herein authors demonstrated the direct vertical integration of Si nanowire arrays into surrounding gate field effect transistors without the need for post-growth nanowire assembly processes. Device fabrication allows Si nanowire channel diameters to be readily reduced to the 5-nm regime [3]. SiNWs are widely used to design effective solar cells [4, 5]. SiNW solar cells based on metal foil receive external quantum efficiency with a maximum value close to 12% at 690 nm [4]. High-quality solar cells were elaborated using a radial junction. The core of the structure was made of a p-doped nanowire grown by CVD using the VLS method [5]. Silicon nanowire arrays are also used as promising scalable anode materials for rechargeable lithium batteries [6, 7].

In order to synthesize nanowires, various methods have been used starting since gas-transport reactions in 1962 [8] and described in more details in [9], and in succeeding years by chemical vapor deposition [10, 11], laser ablation [12], high temperature thermal evaporation [13], molecular or gas-source beam epitaxy [14, 15] and many others based on deposition from solution [16]. Great efforts were applied to understand the growth mechanisms of nanowires, relationship between growth conditions and morphology, composition, crystalline structure, electronic, optical and mechanical properties [17–19], which define fields of nanowires applications [20].

To find technology suitable for realization of these properties, we analyzed various methods of nanowire growth. This analysis as well as results of our previous experiments and simulation of a self-assembling growth of nanowires [21, 22] have shown that the technologies of self-organization are the most promising ones due to simplicity of their realization and low cost. These technologies can be realized in gas-flow reactors of reduced pressure, employing processes of silicon reduction, thermal decomposition, and the processes of gas-transporting reactions as well. The chemical vapor deposition (CVD) method in the open gas transport reactor has become the most popular to form silicon nanowires. In this method, nanowires grow by the vapour-liquid-crystal (VLC) or the vapour-solid-crystal (VSC) mechanisms.

2. Experimental

In this paper, we present results of silicon nanowire growth, which is based on gold enhanced CVD of silicon due to decomposition of silane in gas-flow reactors with hot and cold walls. The used method consists of two main stages. The first one is formation of growth-seeds,
which includes initial refining and treatment of the surface of a substrate, and patterning of the substrate with an array of metal particles (growth-seeds). The second stage consists of growing the nanowires due to decomposition of Si-containing gas molecules at growth-seeds, following transport of rejected silicon atoms through or/and across the growth-seeds and incorporation of Si-atoms into solid phase at the interface or side walls of nanowires.

Particular properties of the growth-seeds, namely: composition, morphology, size distribution, their density and aggregative state under the wire growth conditions, predetermine the respective parameters of the grown nanowires. Despite the fact that, a great deal of experimental researches has been implemented, there is a lack of good understanding of this phenomenon to exert control over important properties of the system, even through it presents the simplest system like gold on silicon substrate.

Therefore, to provide one-dimensional growth of nanowires, a special attention has been paid to initial refining the surface of substrate and methods providing formation of growth-seeds on a substrate. For this purpose, one commonly uses two kinds of methods: (i) deposition of thin metal films and following its annealing or (ii) deposition of previously formed identical nanoparticles. Our previous experience showed that both of the methods result in similar parameters of growth-seeds and the growing NW arrays as well. In the current research, we restricted ourselves by deposition of thin metal films and following annealing of them in a furnace.

Test experiments of silicon nanowire growth were carried out on the produced growth facilities presented in Fig. 1 by using several tens of technological condition sets. The selected conditions were based on our previous experience obtained during joint research with collaborators T. Kamins and S. Sharma in the frame work of the project “Computer Simulation of Stimulated Growth of Silicon Wires on Silicon Substrate” (CRDF).

For all the cases, the growth took place at the overall pressure of 10 mbar in the hydrogen and silane (100:1) flowing at the rate of 100 ml/s. Depending on the temperature and time of both the growth seeds formation and the growth of nanowires, the geometrical characteristics of nanowires were different.

### Table. Technological conditions for the annealing and growth of silicon nanowires and their averaged morphological characteristics.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thickness of the Au deposited film, nm</th>
<th>Temperature of the Au film annealing, °C</th>
<th>Time of the Au film annealing, min</th>
<th>Temperature of NWs growth, °C</th>
<th>Time of NWs growth, min</th>
<th>Medium diameter of the grown SiNWs</th>
<th>Medium length of SiNWs of the grown</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2a)</td>
<td>&lt; 2</td>
<td>560</td>
<td>5</td>
<td>560</td>
<td>10</td>
<td>No growth</td>
<td>~ 15 nm ~ 150 nm</td>
</tr>
<tr>
<td>(2b)</td>
<td>5.0</td>
<td>560</td>
<td>5</td>
<td>580</td>
<td>10</td>
<td>No growth</td>
<td>~ 15 nm ~ 200 nm</td>
</tr>
<tr>
<td>(3a)</td>
<td>4.3</td>
<td>580</td>
<td>5</td>
<td>580</td>
<td>13</td>
<td>~ 15 nm</td>
<td>~ 150 nm</td>
</tr>
<tr>
<td>(3b)</td>
<td>4.4</td>
<td>580</td>
<td>10</td>
<td>580</td>
<td>20</td>
<td>~ 15 nm</td>
<td>~ 200 nm</td>
</tr>
<tr>
<td>(4a)</td>
<td>4.8</td>
<td>620</td>
<td>5</td>
<td>620</td>
<td>30</td>
<td>~ 200 nm</td>
<td>~ 7 µm</td>
</tr>
<tr>
<td>(4b)</td>
<td>2.7</td>
<td>620</td>
<td>5</td>
<td>620</td>
<td>30</td>
<td>~ 150 nm</td>
<td>~ 5 µm</td>
</tr>
<tr>
<td>(5a)</td>
<td>3.5</td>
<td>640</td>
<td>5</td>
<td>640</td>
<td>30</td>
<td>~ 150 nm</td>
<td>~ 50 µm</td>
</tr>
<tr>
<td>(5b)</td>
<td>5.0</td>
<td>640</td>
<td>5</td>
<td>640</td>
<td>30</td>
<td>~ 200 nm</td>
<td>~ 70 µm</td>
</tr>
</tbody>
</table>

3. Results

Technological conditions for the annealing and growth of silicon nanowires and their averaged morphological characteristics are shown in Table.

The crystals do not grow at temperatures below 580 °C and growth times below 15 min (Fig. 2).

Increasing the growth time leads to growing the length of nanowires. On the other hand, the increase in the time of sample annealing results in the increase of crystal diameters (Fig. 3).

Similar conditions of seed formation and growth process lead to close parameters of crystals. As can be

Fig. 3. Increase of the growth time leads to the increase of the nanowire lengths (samples (3a) and (3b), respectively).

Fig. 4. Similar conditions of seed formation and growth process lead to close parameters of crystals (samples (4a) and (4b), respectively).
seen from the results of experiment, a large thickness of the gold film leads to the growth of nanowires having a large length and diameter (Fig. 4 and Fig. 5).

4. Conclusion

In this paper, we have presented results of silicon nanowire growth, which is based on gold enhanced CVD technology in gas-flow reactors of reduced pressure. The used method consists of two main stages: formation of growth-seeds and growth of nanowires due to decomposition of Si-containing gas molecules at growth-seeds.

Particular properties of the growth-seeds, namely: composition, morphology, size distribution, their density and aggregation state under the wire growth conditions, predetermine the respective parameters of the grown nanowires. Therefore, to provide one-dimensional growth of nanowires, special attention has to be paid to initial refining the surface of substrate and methods providing formation of growth-seeds on a substrate.

For all the cases, the growth took place at the overall pressure of 10 mbar, proportion of hydrogen and silane 100:1 and the flux of gas mixture 100 ml/s. Depending on the temperature and time of growth-seeds formation and the growth of nanowires, the geometrical characteristics of nanowires were different. We carried out test experiments on growing silicon nanowires using several tens of sets of technological conditions.

As a result of the performed experiments, we know how to grow the silicon nanowires with before desired morphology. The study of optimal growing regimes creates prerequisites for the industrial production of SiNWs.

References


Authors and CV

Klimovskaya A.I. Doctor of Sciences in Physics and Mathematics, Leading Researcher at the Department of Ion Beam Engineering, V. Lashkaryov Institute of Semiconductor Physics, NAS of Ukraine. The area of scientific interests includes semiconductor and dielectric science, surface science, biophysics, crystal growth, nanotechnology, nanoelectronics, optoelectronics, surface science, hot electrons and intervalley electron redistribution near silicon surface; magnetic properties of surface layers near quantum limit; current and/or voltage instability in MOS-structures; femtosecond optical pulse generation; intelligent microsensors of temperature, humidity, light intensity, micro-movement and DC-AC micro-converters; magnetic sensors for high power cryogenic turbo generators operating at extreme temperatures, vibration and rotation loads; magnetic sensors of short magnetic pulses and for diagnostic space distribution of very small magnetic fields; growth silicon and silicon/germanium wires and their application for regeneration of nervous tissue.

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