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Self-consistent modeling of MBE self-catalyzed GaAs nanowire growth

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Abstract: Self-catalyzed GaAs nanowires are synthesized by molecular beam epitaxy at various arsenic fluxes and growth temperatures. The growth of GaAs nanowires is simulated considering the kinetics of material transport inside the catalyst droplet. The re-evaporation coefficient of arsenic is estimated for the given growth conditions. Calculated nanowire growth rate is in satisfactory agreement with the experimental data.

1. Introduction

Epitaxial arrays of III-V semiconductor nanowires (NWs) are a promising material for modern optoelectronic and electronic technologies. Particularly, GaAs NWs can be used for fabrication of novel photovoltaic devices [1]. When forming NW by the vapor-liquid-solid (VLS) mechanism, the most extensive possibilities for controlling the morphology of NWs arrays are opened when using catalytic particles of foreign materials (usually Au droplets pre-deposited on the growth substrate). However, the catalyst material can be embedded into the lattice of growing NWs, resulting in deep-level defects that reduce the lifetime of charge carriers and increase the probability of non-radiative recombination. The NW formation by the self-catalytic mechanism [1], when using a group III element of the NWs itself as a catalyst (namely, gallium for GaAs), excludes contamination of III-V compounds, but imposes restrictions on the range of possible growth parameters and, as a result, the morphology of the formed NWs arrays [2].

In this paper, the growth rate of self-catalyzed GaAs NWs is found at different arsenic fluxes and temperatures both experimentally and theoretically. The model proposed in [3] within the “As-only” approach [4] is used for the theoretical assessment of the GaAs NWs growth rate. The value of the re-evaporation coefficient of arsenic is calculated for the given growth conditions by fitting the experimental growth rate.

2. Theory

In the model [3], the nucleation-limited growth rate of NWs which is given by the Zeldovich nucleation rate times the surface area of the top NW facet is considered. It is assumed that the growth rate of 2D islands on the NW top facet is limited by the diffusion flux of arsenic inside the gallium droplet. The simplified geometry of the system is presented in the inset in figure 1b. The growth



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modeling is performed by two steps. Firstly, for each monolayer, the duration of the droplet refilling stage t_s is calculated. The time t_s is estimated by using the expression for the steady-state nucleation rate:

$$t_s \approx (t + \tau_N(t))_{\min}. \quad (1)$$

Here $\tau_N(t)$ is the time between two consecutive nucleation events calculated at the moment of time t , $\tau_N = 1/(\pi R_0^2 I)$, where I is the Zeldovich nucleation rate, R_0 is the NW radius. Self-consistent modeling is achieved by considering the material balance equation for arsenic in the droplet:

$$\frac{dN_{As}}{dt} = J_{As}^{dir} + J_{As}^{re} - J_{As}^{des}, \quad (2)$$

where N_{As} is the number of arsenic atoms in the droplet; J_{As}^{dir} and J_{As}^{re} are the direct and re-evaporated arsenic flux in the vapor phase, J_{As}^{des} is the desorption arsenic flux. Following the approach [4], the sum of the direct and re-evaporated arsenic fluxes is given by the formula:

$$J_{As}^{dir} + J_{As}^{re} = (1 + \varepsilon_{As}) j_{As}^{dir} S, \quad (3)$$

where ε_{As} is the arsenic re-evaporation coefficient, j_{As}^{dir} is the arsenic flux density, S is the cross section of the droplet. Secondly, the lateral growth of the GaAs nucleus (GaAs island) is modeled. The incorporation flux J_{As}^{inc} should be introduced in the material balance equation:

$$\frac{dN_{As}}{dt} = J_{As}^{dir} + J_{As}^{re} - J_{As}^{des} - J_{As}^{inc}. \quad (4)$$

The island growth rate is determined by the equation:

$$\frac{dR}{dt} = \frac{\Omega}{2\pi h R} J_{As}^{inc}, \quad (5)$$

where Ω is the volume of GaAs pair in solid state, h is the GaAs monolayer height, R is the island radius. The analytical expression derived in [3] is used for the incorporation flux. This expression was obtained by solving a steady-state diffusion problem, and it was assumed that the island has the shape of a disk. It was supposed that the equilibrium concentration of arsenic is maintained at the island edge. Also, the attachment rate of arsenic atoms to the critical island introduced in the nucleation rate I is derived in [3].

3. Experiment

Epitaxial GaAs NWs arrays were synthesized on the vicinal Si (111) substrate with a 4° miscut orientation in [112̄] direction using the MBE (molecular beam epitaxy) Veeco GEN-III machine with Ga effusion cell and valved arsenide cracker for As₄ molecular flux. Before loading into the MBE chamber, the silicon substrates were cleaned using a modified RCA cleaning and Shiraki etch procedures [5, 6]. Oxidation was carried out by boiling in “base piranha” mixture or RCA-1 cleanser (aqueous solution of ammonia and hydrogen peroxide NH₄OH:H₂O₂:H₂O with the volume ratio of 1:1:3) [7]. The substrate temperature was monitored with a thermocouple calibrated using the silicon oxide decomposition and Si (111) 7x7 to 1x1 phase transition temperatures [8]. Bayard-Alpert vacuum gauge was used to control molecular fluxes of groups III and V and BEP (beam equivalent pressure).

Silicon substrates with oxide were preliminarily degassed in the intro and buffer chambers, and then annealed in the growth chamber, at a temperature lower than the surface oxide decomposition temperature by 20 degrees, to create defects in the oxide layer, which are necessary for the growth of vertical NW. Thereafter, before the growth process, gallium was pre-deposited, which self-organized

into droplets. The growth was carried out at the temperatures of 570°C and 600°C (for 10 min and 30 min, respectively) and at the As₄ molecular fluxes of 1.5×10^{-6} Torr and 3×10^{-6} Torr. In this case, the gallium flux was constant (1.25×10^{-7} Torr) which corresponds to the growth rate of planar GaAs 313 nm/h.

The morphology of synthesized GaAs NWs was studied with scanning-electron microscope (SEM) (Zeiss SUPRA 25-30-63).

4. Results and discussion

The re-evaporation coefficient of arsenic ε_{As} that characterizes the arsenic evaporation flux from the substrate and the NW sidewalls is used as the sole fitting parameter. The estimated value of ε_{As} equals 6.5 (two times larger than that obtained in [4]). The value of ε_{As} depends strongly on the surface density of NWs and their morphology. Also, the following value of the conversion factor η connecting the arsenic BEP and the arsenic flux density ($j_{As}^{dir} = \eta \times BEP$) is found $1.28 \times 10^{24} \text{ m}^{-2} \text{ s}^{-1} \text{ Torr}^{-1}$. This value is about two times smaller than that used by Glas et al. [4], because it depends on the configuration of MBE machine. However, the value of the sum $J_{As}^{dir} + J_{As}^{re}$ (equation (3)) is in agreement with the results of the paper [4]. The calculation is performed at the interphase energy of the GaAs island side facet (0.309 J m^{-2}) [3]. The dependences of the NW growth rate on the group V flux calculated at different growth temperatures are shown in figure 1b.

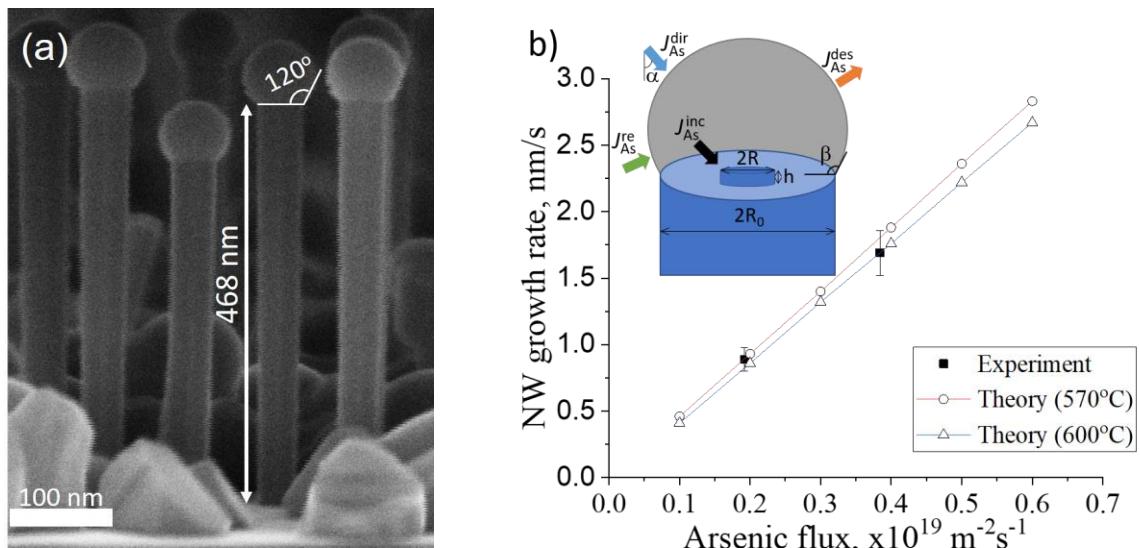


Figure 1. a) Cross-sectional SEM image of GaAs NW array. b) Calculated (lines) and experimental (dots) dependencies of NW growth rate on arsenic flux (red and blue lines are dependencies simulated at 570°C and 600°C, respectively). Schematic representation of self-catalyzed VLS growth of GaAs NW is shown in the inset. The values of NW radius $R_0 = 51 \text{ nm}$, contact angle $\beta = 120^\circ$ and incident angle of the arsenic flux $\alpha = 30^\circ$ are used in computation.

Table 1. NW length calculated within the model [3] and measured in the experiment.

Sample	Calculated length (μm)	Measured length (μm)
S1	3.042	2.998 ± 0.3
S2	0.534	0.502 ± 0.04

It is found that the growth rate obtained in the model is consistent with the real growth rate of synthesized GaAs NWs with an accuracy of about 80-85 percent.

5. Conclusions

In summary, self-catalyzed GaAs NW formation was studied both experimentally and theoretically. The values of the growth rate of self-catalyzed GaAs NWs predicted by the model are in a good agreement with the experimental data. The re-evaporation coefficient of arsenic was used as a sole fitting parameter.

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