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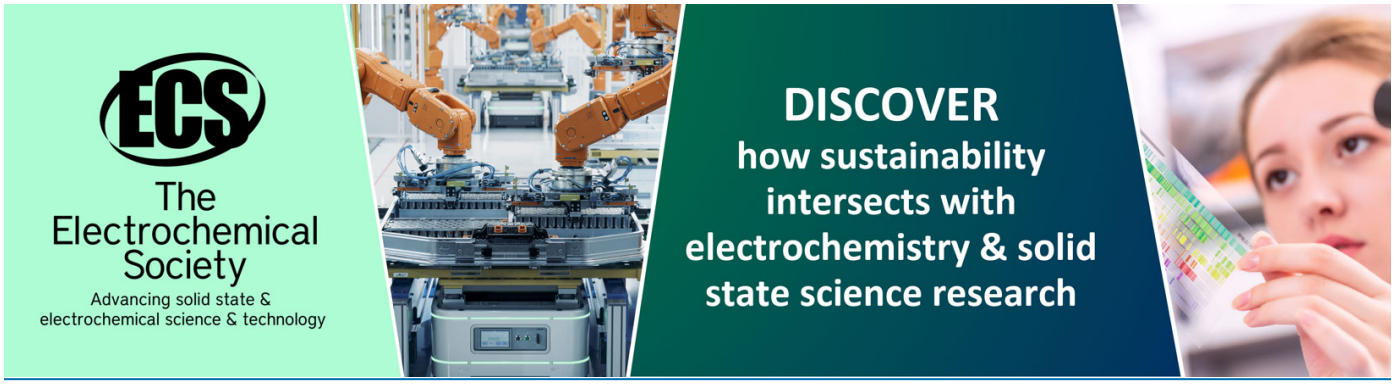

Self-induced GaN nanowire growth: surface density determination

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Abstract. A new numerical approach for the determination of the GaN nanowire surface density on an AlN/Si substrate as a function of the growth time and gallium flux is presented. Within this approach, the GaN island solid-like coalescence and island-nanowire transition are modeled by the Monte-Carlo method. We show the importance of taking into consideration the island coalescence for explaining that the maximum of GaN island surface density is several times larger than the maximum of GaN nanowire surface density. Also, we find that the nanowire surface density decreases with an increase of the gallium flux.

1. Introduction

The wide bandgap and good transport properties make gallium nitride ideal candidates for production of high power electronic and optoelectronic devices [1]. The GaN film growth on substrates with lattice mismatch larger than 2% [2] results in the formation of large amount of dislocations in the GaN layer. The GaN nanowire-based devices can solve this problem. Nanowires (NWs) grow on substrates with a large lattice mismatch without dislocation formation because of a specific elastic stress relaxation mechanism [2]. The present paper is devoted to the modeling of self-induced GaN NW formation (catalyst-free growth) on AlN/Si substrate. The self-induced NW growth enables to avoid an unwonted contamination of the NWs by the catalyst material. Despite the semiconductor NW growth have been studied intensively for the last decade [2, 3] the mechanisms of self-induced NW growth are still far from being fully understood. Consonni [4] distinguishes the three growth stages of self-induced GaN NWs on AlN/Si substrate. The nucleation stage of GaN islands (or the incubation period) is a time interval since the onset of material deposition until the GaN islands appear on the substrate. The incubation period can vary in a typical range of several tens of seconds to several tens of minutes. During the transition stage, the islands grow and change their form (presumably, in the following sequence: cap-shaped islands, truncated pyramid-shaped islands, full pyramid-shaped islands) as a result of elastic stress relaxation. The GaN islands transform into the NW (column-shaped islands) when the island lateral size reaches the critical size that is determined by the onset of plastic relaxation. Finally, an intensive vertical growth of NWs occurs during the NW growth stage. In the work [5], we modeled the nucleation stage of GaN islands and found the size distribution function of islands using the model of the growth via the Stranski-Krastanov mechanism [6, 7]. The island and NW surface density was calculated assuming that any island reached the critical size transforms into NW and neglecting the island coalescence. In this paper, we use a numerical approach wherein the island solid-like coalescence is modeled by the Monte-Carlo method. As a result, the accuracy of the surface density calculation is essentially improved.

2. Model

The initial size distribution function of GaN islands has the form of [5]

$$f(\rho,t) = cn\exp\left(cx(\rho,t) - \exp(cx(\rho,t))\right). \quad (1)$$

Here $\rho = \left(r/\Omega^{1/3}\alpha\right)^2$ is the invariant island size, r is the island base radius, Ω is volume per GaN pair, $\alpha = \left(3/\pi\eta(4\eta^2+3)\right)^{1/3}$ is the geometric factor for the spherical cap-shaped island, η is the island aspect ratio, n is the maximal surface density of island after the nucleation stage, c is a coefficient that characterizes the distribution function dispersion, $x(\rho,t) = \rho_0(t) - \rho$, $\rho_0(t)$ is the island size that corresponds to the distribution function maximum.

We model the island formation on the rectangular substrate with the side $L = \left(N/n\right)^{1/2}$, N is the maximal number of islands in our model. To perform the Monte-Carlo modeling, the island size distribution function is discretized and the following rules are used. At time $t = 0$, the island distribution function is completely in the region of negative island sizes. At each step of the modeling, the growth rate of the invariant sizes of islands and NWs, $\mathrm{d}\rho/\mathrm{d}t$ and $\mathrm{d}\rho_1/\mathrm{d}t$ respectively, are found from the material balance equation that is written in terms of supersaturation for islands and for NWs [5]

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \frac{1}{\tau}\left(\frac{V\chi t}{h_{\mathrm{eq}}} - \frac{S}{L^2 h_{\mathrm{eq}}}\left(N_{\mathrm{ISL}} + N_{\mathrm{NW}}\right)\right), \quad (2)$$

$$\frac{\mathrm{d}\rho_1}{\mathrm{d}t} = \frac{1}{\tau_1}\left(\frac{V\chi t}{h_{\mathrm{eq1}}} - \frac{S}{L^2 h_{\mathrm{eq1}}}\left(N_{\mathrm{ISL}} + N_{\mathrm{NW}}\right)\right). \quad (3)$$

Here ρ_1 is the invariant size for column-shaped islands (i.e. NWs) [5], the first term in the brackets in the right hand side of equations (2) and (3) corresponds to the total amount of material on the substrate, the second term corresponds to the total amount of material in the islands and NWs, V is the Ga flux (in ML/s), χ is the sticking coefficient, t is the growth time (the wetting layer thickness equals to the equilibrium thickness at $t = 0$), τ, h_{eq} are the diffusion time and the equilibrium wetting layer thickness for the islands and NWs (index 1 corresponds to NWs), S is the surface area per GaN pair, N_{ISL} and N_{NW} are the total amount of GaN pairs in the islands and NWs. Using the following formulas, we can find the lateral size of islands and the height of NWs: $r = \alpha\Omega^{1/3}\rho^{1/2}$ and $L = \Omega/(\pi r_{\mathrm{C}}^2)\rho_1$, respectively , where r_{C} is the critical island size. As soon as the island size ρ becomes positive, the island (i.e. a circle with radius r) appears in a random place of the substrate (figure 1). If an island nucleates inside other island or NW this island does not grow. If an island reaches the critical island size r_{C} before collision with other island or NW the island becomes NW. Otherwise, the island-NW transition does not occur. If two or more islands collide we count these islands as one. When an island collides with other island or a NW the islands and NW continue growth as if there were no collisions. The NW radius is a constant in the model and equals to r_{C}. The suggested scheme allows the modeling of the GaN NW formation. Figure 1 shows a typical view of the substrate during different growth stages of NWs. One can see in figure 1 multiple collisions of islands that decrease drastically the island surface density. To validate our model, we compare the island and NWs surface density dependence on time with the experimental data [8]. The parameters of the model used in this computation are: $\eta = 0.11$, $\rho_{\mathrm{C}} = 500$ ($r_{\mathrm{C}} \approx 9$ nm that two times smaller than in [4]), $V = 0.09$ ML/s, $\chi = 0.2$, $h_{\mathrm{eq}} = 3$ ML, $\tau = 6.5\times10^{-2}$ s, $h_{\mathrm{eq1}} = 3$ ML, $\tau_1 = 1.0\times10^{-5}$ s, $c = 0.013$, $n = 1300\ \mu m^{-2}$. The value of V and h_{eq} is known from the experiment [8], the value of η was

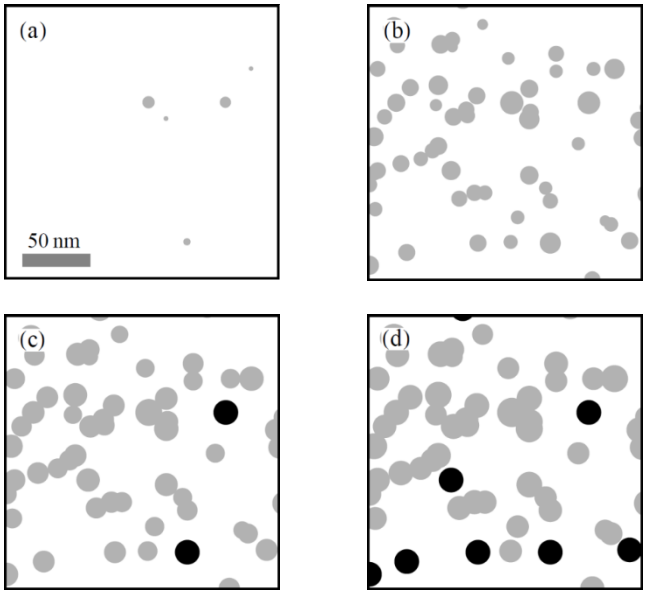

Figure 1. The substrate during modeling at $t = 70$ s (a), $t = 110$ s (b), $t = 150$ s (c), $t = 1300$ s (d). The gray circles are the GaN islands, the black circles are the GaN NWs. The parameters used are described in the text.

calculated in the work [5]. The value of the maximal island density n is higher than in the experiment [8] (about $950\ \mu m^{-2}$) because, as we assume, very small nuclei were not detected in the experiment before they reached some size and collided with each other. In fact, n is a fitting parameter of the model. The parameters χ, τ and c are found using the method [7] and depends on the choice of n. Thus, the fitting parameters of the model are: r_C, n, h_{eq1} and τ_1. Figure 2 shows a good agreement of the theoretical and experimental dependences.

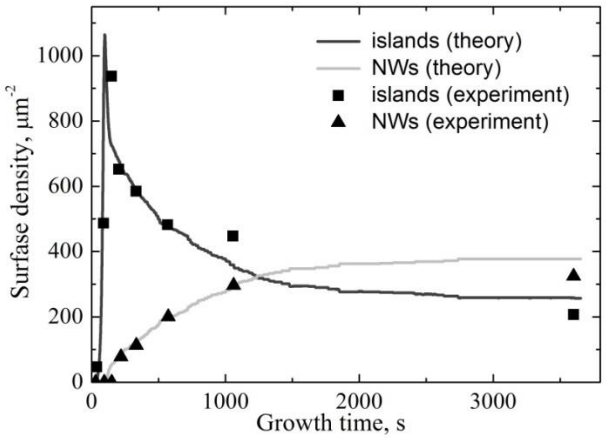

Figure 2. The GaN island and GaN NW surface density versus the growth time. The parameters used to obtain the theoretical curves are described in the text.

On the basis of the modeling, we can make the following notes regarding the NW formation. Firstly, the nucleation stage of GaN islands is short (t = 0-150 s). Secondly, at the very onset of the transition stage (t = 150 s) a large decrease of the island density occurs as a result of the multiple collisions of islands with the size lower than r_C. Therefore the maximal island density is about three times higher than the maximal surface density of NWs. Thirdly, as the transient stage (t = 150-1300 s) starts, the island growth rate decreases drastically and can be even negative (the island evaporation) since the NW growth rate is much higher than the island growth rate ($\tau >> \tau_1$). Fourthly, at t > 1300 s (the NW growth stage), almost all islands either transform into NWs or collide therefore the island and nanowire surface density change insufficiently.

It is noteworthy that the NW coalescence process [9] and the shadowing effect [10] are excluded from our consideration. Although these effects play an important role when the NW height and NW density is large enough. Also, we neglect the island shape change produced by the elastic stress relaxation assuming that the island growth regime changes instantly at the island-NW transition only [5].

Our approach allows the estimation of the NW surface density for different gallium fluxes. For this purpose, we calculate the island size distribution function for different gallium fluxes using the method [7]. Then, we model the NW growth with different initial distribution function. Although the maximal island density increases with an increase of material flux [7], the NW surface density decreases (figure 3) as a result of multiple island collisions that decreases several times the amount of

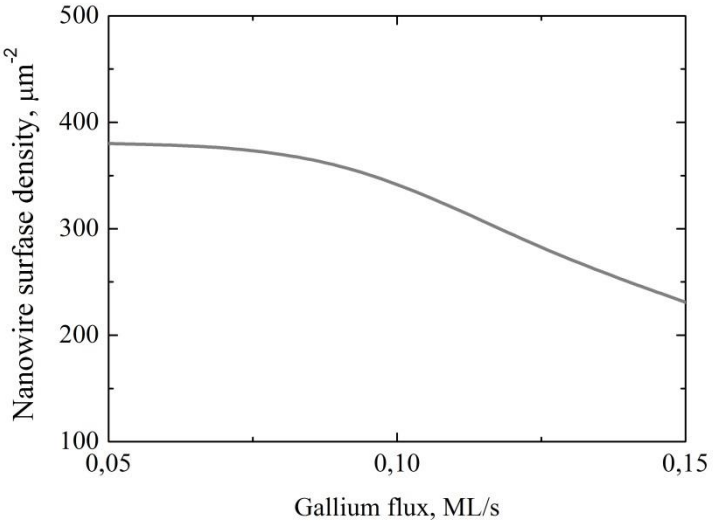

Figure 3. The NW surface density versus the gallium flux.

single islands. It should be noted that the NW surface density is an increasing function of material flux in the model without island collisions [5].

3. Conclusion

A new numerical approach for the determination of the GaN NW surface density on an AlN/Si substrate as a function of the growth time and gallium flux is presented. To model the GaN NW growth, we use the initial size distribution function of GaN islands calculated elsewhere. The island and NW growth rate are found from the material balance equation. The GaN island solid-like coalescence and island-NW transition are modeled by the Monte-Carlo method assuming several rules: if an island reaches the critical island size before collision with other island or NW the island becomes NW, otherwise, the island-NW transition does not occur; if two or more islands collide we count these islands as one; if an island nucleates inside other island or NW this island does not grow;

when an island collides with other island or a NW the islands and NW continue growth as if there were no collisions. We show the importance of taking into consideration the island coalescence for explaining that the maximum of GaN island surface density is several times larger than the maximum of GaN NW surface density. Also, we find that the NW surface density decreases with an increase of the gallium flux as a result of multiple island collisions that decreases several times the amount of single islands.

Acknowledgments
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