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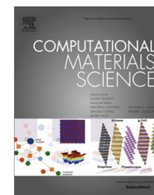

Stochastic model and simulation of growth and coalescence of spontaneously formed GaN nanowires in molecular beam epitaxy

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ABSTRACT

The stochastic model of the growth of an ensemble of GaN nanowires (NW) in a plasma-assisted molecular beam epitaxy suggested in our recent paper (Sabelfeld and Kablukova, 2016) is here further developed to include coalescence caused by bundling of nanowires. Moreover, in the extended model the Ga and N atoms are simulated separately to mimic nucleation of stable GaN islands which then start to grow in one direction with a fixed diameter. The bundling phenomenon is driven by the gain of surface energy at the expense of the elastic energy of bending and becomes energetically favorable once the nanowires exceed a certain critical length as found and described in our paper (Kaganer et al., 2016). The simulation model is based on a probabilistic description of surface diffusion, and takes into account the shading, multiple rescattering of atoms, and their survival probability. The model is implemented in a form of a direct simulation Monte Carlo algorithm. We present a comprehensive analysis of the kinetics of NW growth from an initial height distribution around tens of nanometers to NW heights up to several thousands nanometers which corresponds to the growth time in physical growth experiments about of 3–4 h. We compare the simulation results with our recently developed model without bundling and experimental results as well which show a good agreement. The series of simulations have basically confirmed the remarkable time evolution of the nanowire height distribution, namely, that under some conditions, now described more precisely, the NW height distribution is self-preserving and may become even narrower independent of the form of the initial NW height distribution. This phenomenon is explained by the multiple rescattering of atoms between the NW surfaces.

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1. Introduction

In the synthesis of nanowires in a plasma-assisted molecular beam epitaxy (PA-MBE) the GaN nanowires are formed spontaneously on different substrates under appropriate conditions like N excess and elevated temperatures [1]. The spontaneous formation of GaN nanowires in PA-MBE occurs without any metal particle on their top, in contrast to the vapor-liquid-solid (VLS) growth approach. The NW growth by PA-MBE technique has initiated exciting fundamental research and opened a way to innovative electronic devices (e.g., see [2–4]).

The mechanism of spontaneous formation of GaN nanowires on bare Si(111) substrates have been intensively studied (e.g., see [5–7]). In [8] we have studied experimentally and partly theoretically the axial and radial growth of GaN nanowires upon a variation of the Ga flux during molecular beam epitaxial growth. An increase in the Ga flux promotes radial growth without affecting the axial growth rate. In contrast, a decrease in the Ga flux reduces the axial growth rate without any change in the radius.

In [4] we have studied the time evolution of GaN nanowires grown on Si(111) substrates by PA-MBE technique under the temperature of 800 °C. In these experimental study we have observed a NW height equilibration phenomenon where the height distribution was self-preserving in time. To explain this unexpected behavior, we have suggested in [4] a phenomenological stochastic model of NW growth based on a system of stochastic differential equations. This growth model of self-induced GaN nanowires involves the exchange of Ga atoms between nanowires: Ga atoms desorbed from the NW sidewalls readsorb on neighboring nanowires. This process favors the growth of shorter nanowires and gives rise to a narrow nanowire height distribution during the late stages of growth. The model however involves an unknown function responsible for the scattering of atoms between the neighboring

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nanowires. This model being phenomenological, could not explain why the NW height distribution in some growth experiments was not only self-preserving, but even narrowing with time. To explain this narrowing height distribution during the NW growth and to validate the phenomenological model [4], we have developed in [9] a direct Monte Carlo algorithm which simulates the bottom-up synthesis of a large ensemble of GaN nanowires. The model is based on probabilistic description of the surface diffusion, shading, multiple scattering of atoms on the neighboring nanowires, and survival probabilities. In recent study [11] we have investigated the nucleation, growth and coalescence of the GaN nanowires by combining the statistical analysis of scanning electron micrographs with Monte Carlo growth models. There we have found that shortly after the nucleation stage, nanowire radial growth exponentially decreases and becomes negligible, while during the further growth, the close standing nanowires coalesce by bundling. The coalescence is driven by the gain of surface energy at the expense of the elastic energy of bendling and becomes energeticall favorable once the nanowires exceed certain critical length [11,12].

This phenomenon has a considerable impact on the NW growth dynamics, therefore, we have included it in the Monte Carlo simulation model in the present paper. We have considered two regimes of bundling process: in the first version each nanowire may coalescence only once. In the second version, the coalescence may form a conglomerate of 5–6 nanowires. Another important feature of the spontaneous formation of GaN nanowires in PA-MBE is the different impact of N and Ga atoms on the radial and axial growth rate. It was not taken into account in our previous model [9], and is now properly developed in the extended version of the model developed in the present paper.

The paper is organized as follows. In Section 2 we describe the stochastic models developed: Section 2.1 outlines the basic stochastic growth model and main steps of the simulation algorithm where the bandling is not included, and N-rich conditions are assumed. In Section 2.2 we present the coalescence of nanowires and explain how this phenomenon is incorporated into the growth model and simulation algorithm. In Sections 2.3 and 2.4 we extend the model to include the detailed and separate simulation of Ga and N atoms which enables to simulate the appearance of new nanowire nuclei on the substrate under a limited supply of N atoms. This comes from the fact that N atoms have a small diffusion length both on the substrate and on the sidewall facets of GaN nanowires and desorb as inert N_2 molecules. Therefore, the amount of reactive N atoms reaching the substrate is negligible, which in turn prevents further nucleation once a shading sets in. In Section 3 all simulation results are presented and discussed.

2. The NW growth simulation algorithm

2.1. Outline of the basic model and simulation algorithm

Let us start with the setting of the basic model without coalescence and conditioned generation of new NW nuclei. The description of the basic growth model we use for simulation of the GaN nanowires is close to that presented in our paper [9]. The substrate surface is taken in the form of a circle. The flux of adatoms is inclined with respect to the substrate normal, the atoms impinge on both the top and side surfaces of the nanowires. The nanowire is considered as a circular cylinder. Rotation of the substrate about its axis provides irradiation of the side surfaces from all directions.

The parameters of the model were chosen in accordance with the NW growth carried out under the experimental conditions which we have described in [4]. The main featues can be described briefly as follows. A set of GaN nanowire samples was grown on Si (111) substrates by plasma-assisted molecular beam epitaxy in the self-induced way. All samples were grown at the same temperature of 800 °C. The Ga and N fluxes (calibrated as growth rates of GaN films grown under Ga- and N-rich conditions) were 4 nm/min and 18 nm/min, respectively. The growth time was the only parameter that varied from one sample to another, in the range 15–120 min. The time was counted starting from the shutter opening, so that the nucleation time is included.

In the basic model, we start with generation of a set of small nanowires of radii r_i and heights h_i which correspond to the experimental results to the time instant 30 min as suggested in [4,9]. Each nanowire in this set is considered as a circular cylinder of finite height. The NW radius r and height h are random having log-normal distributions

$$p(x) = \frac{1}{x\sigma\sqrt{2\pi}} e^{-(\ln x - \mu)^2/2\sigma^2} \tag{2.1}$$

with a mean radius $\mathbf{E}r$ and mean height $\mathbf{E}h$, and variances $\mathbf{V}r$ and $\mathbf{V}h$. The mean and variance of the lognormal distribution are related with the mean μ and variance σ^2 of a normal distribution by

$$\mathbf{E}r = e^{\mu+\sigma^2/2},\ \mathbf{V}r = \left(e^{\sigma^2} - 1\right)e^{2\mu+\sigma^2}.$$

Periodic conditions are supposed on the boundary of the substrate. The generation of the initial NW distribution on the substrate is carried out as follows. The positions of the cylinders on the substrate are generated randomly and uniformly on the substrate in accordance with the prescribed coverage ΔS. The coverage is defined as the ratio of the total area of the bottoms (discs) of all nanowires to the area of the substrate. The radius of the substrate R_{sb} and the coverage ΔS are related by $R_{sb} = \mathbf{E}r\sqrt{N/\Delta S}$ where N is the number of nanowires, and $\mathbf{E}r$ is the mean value of the NW radii. The coordinates of the centers of NW's bottoms are sampled now using the following algorithm. We introduce a mesh with the nodes formed by the centers of equal closely-packed discs in circule of unit radii. We used the coordinates for a set of discs for different values of N from [13], and then the desired mesh is obtained by scaling for the substrate of radius R_{sb}. The nanowire position indexed by an integer k is then sampled in two steps.

1. Choose at random the index k_{nd} of a free node of the mesh as an integer number with uniform distribution among N nodes.
2. In the sampled disc with the index k_{nd} sample the coordinates of the NW's center according the uniform distribution in this disc. The sampled discs (the bottoms of the nanowires) should satisfy the following condition: the distance between any two nanowires cannot be less than a prescribed value ε:

$$\sqrt{(x_k - x_i)^2 + (y_k - y_i)^2} > R_k + R_i + \varepsilon. \tag{2.2}$$

If for some node k_{nd} this condition cannot be satisfied, it is assumed that this node is empty. The value ε is taken proportional to the diffusion length.

The diffusion length for Ga on the NW's side surface is defined by $L = \sqrt{D\tau}$, which is the mean distance of the diffusing Ga atom before its desorption, and τ is its mean diffusion time. Taking $\tau = 1$, the desorption coefficient is defined by $D = L^2$. In what follows, we denote by θ the angle between the normal vector to the substrate and the direction of the incident flux of atoms.

The simulation of the NW growth is based on tracking of atom trajectories from the source, followed by the intersection with the top or side surfaces of the nanowire, diffusion over the side surface, desorption and rescattering from one nanowire and again, possible intersection with one of other nanowires.

The adatom's diffusion over the NW side surface is simulated as follows. First we simulate the diffusion along the vertical

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