08.2

Growth kinetics of III-V nanomembranes influenced by the re-emitted flux of group III species

© V.G. Dubrovskii

St. Petersburg State University, St. Petersburg, Russia E-mail: dubrovskii@mail.ioffe.ru

Received April 12, 2023 Revised May 5, 2023 Accepted May 5, 2023

A growth equation for III–V nanomembranes (NMs) fabricated on patterned substrates with one-dimensional array of trenches is derived considering the re-emitted flux of group V species. It is shown that the NM height increases with the pitch of trenches and for narrower trenches. Overall, the growth kinetics and the resulting morphology of III-V NMs on reflecting substrates is very different from those on adsorbing substrates, which should be carefully accounted for in the growth experiments.

Keywords: III-V compound semiconductors, nanomembranes, re-emitted flux, growth rate, modeling.

DOI: 10.61011/TPL.2023.07.56439.19590

Nanomembranes (NMs) of III-V (III-V NMs) semiconductor compounds and heterostructures based on them [1-7], which are grown by selective area epitaxy (SAE) on various substrates with one-dimensional (1D) trenches in an oxide mask, are used in fundamental research into low-temperature transport and have a wide range of promising applications in opto- and nanoelectronics. An NM growth theory needs to be developed in order to control and adjust the morphology of NM arrays directly in the process of SAE. Current models of the NM formation process [7,8] are rather poorly developed compared to the growth theory of III-V nanowires (NWs) obtained by SAE [2,9] or in the "vapor-liquid-solid" growth mode [10–14]. The aim of the present study is to extend the theoretical model of growth of an individual III-V NM, which was proposed in [8], to include an ordered array of NMs with a preset distance between them (pitch P) and the re-emitted (or desorbed) flux of group III species reflected from the mask surface and adjacent NWs [14-17] taken into account.

Since NM width W and its height H measured from the oxide mask surface are much smaller than the diffusion length of group III adatoms on the top and side faces of a rectangular NM, the general expression for the height growth rate of an individual NM [8] under enrichment in group V species is

$$\left(\frac{dH}{dt}\right)_{dir} = \upsilon \left(\Phi + \frac{2H}{W}\varepsilon\right). \tag{1}$$

Here, index *dir* denotes the direct flux without re-emission (this effect will be considered below). Quantity v is the rate of deposition of group III atoms from the gas phase (nm/s), $\Phi \leq 1$ is the coefficient characterizing desorption from the top NM face (thus, $v\Phi$ is the two-dimensional (2D) growth rate on this surface), and $\varepsilon \leq 1$ is the coefficient of "transmission" of adatoms of a group III element that

migrate from the side faces of an NM to its top face. Quantity ε is related to the rate constants of transitions of adatoms from the side faces of an NM to its top face and back and to the lifetimes of adatoms on these faces [7,8,13]. It is assumed in what follows that $\varepsilon > 0$. Growth rate (1) of a 1D NM is equivalent in this case to the growth rate of vertical NWs [9–13], wherein quantity $2H\varepsilon/W$ characterizes the diffusion contribution. Diffusion from the mask surface is neglected in expression (1) [7,8,13], which is valid for vapor-phase epitaxy (VPE) [16]. Selective area growth by molecular-beam epitaxy (MBE) is characterized in a similar way, but one needs to take the directional nature of a gas flux at MBE into account [15].

It is evident that a dependence on pitch P is lacking in model (1). It is assumed in what follows that re-emission of group III element atoms is the key mechanism of material exchange between an NM array and the mask surface. These atoms are first reflected from the surface and then get deposited onto the NM surface [14–17]. It follows from material balance considerations that desorption flux v_{des} satisfies equation

$$v = \frac{1}{P} \frac{d}{dt} (WH)_{dir} + v_{des}.$$
 (2)

The length of a 1D NM array is considered to be infinite, and edge effects are neglected. In accordance with (2), overall flux v to the surface is equal to the sum of a change in the NM array volume due to the direct flux and the desorption flux from all surfaces [15–17]. Assuming that NW width W is constant and using (1), we obtain the following from (2):

$$v_{des} = v \left(1 - \frac{P_{nm}}{P} \right), \qquad P_{nm} = W\Phi + 2H\varepsilon \leqslant P,$$
$$v_{des} = 0, P_{nm} > P. \qquad (3)$$

It is taken into account here that the desorption flux goes to zero at the time point when effective collection length P_{nm} of group III adatoms becomes equal to pitch P. At $P_{nm} > P$, an NM array consumes completely the gas flux to the surface. The overall NM growth rate in the vertical direction is equal to the sum of rates of growth from direct and reflected fluxes:

$$\frac{dH}{dt} = \left(\frac{dH}{dt}\right)_{dir} + \left(\frac{dH}{dt}\right)_{des}, \ \left(\frac{dH}{dt}\right)_{des} = v_{des}\frac{P_{nm}}{W}.$$
(4)

It is taken into account in expression $(dH/dt)_{des}$ for the rate of growth from the reflected flux that desorbed group III element atoms raise the density of gas that surrounds uniformly the NM array in VPE. It then follows from (1) and (3) that

$$\frac{dH}{dt} = v \frac{P_{nm}}{W} \left(2 - \frac{P_{nm}}{P} \right), \qquad P_{nm} \leqslant P,$$
$$\frac{dH}{dt} = v \frac{P}{W}, \qquad P_{nm} > P, \qquad (5)$$

where the effective collection length is defined in (3). Thus, the rate of vertical MNM growth first increases linearly with *H* and then reaches saturation at level vP/W, which corresponds to the gas flux to one NM. Equation (5) at $P_{nm} \leq P$ may be made dimensionless:

$$\frac{dy}{dx} = y(2 - y),\tag{6}$$

where $y = P_{nm}/P \le 1$ and $x = 2\varepsilon vt/W$. The integration of (6) with an initial condition corresponding to H(t = 0) = 0 yields the key result of the present study:

$$H = \frac{P}{\varepsilon} \frac{1}{1 + (2P/W\Phi - 1)\exp(-4\varepsilon vt/W)} - \frac{W\Phi}{2\varepsilon},$$
$$H \leqslant \frac{P - W}{2\varepsilon},$$
$$H = \frac{P - W}{2\varepsilon} + \frac{P}{W}v(t - t_*),$$
(7)

where t_* is the time point when the NM height reaches critical value $(P - W)/2\varepsilon$.

According to (7), the NM growth law at the first stage is more complex than an exponential one. An exponential height increase follows from (1) with the re-emitted flux neglected. *H* becomes a linear function of time as soon as a critical height is achieved. This growth pattern resembles the temporal evolution of vertical NWs without radial growth [15,16], but has different dependences on *W* and *P* that are typical of 1D geometry. Figure 1 shows the dependences of NM array height *H* on effective thickness at fixed NM width W = 160 nm and pitch *P* varying from 400 to 1200 nm. Figure 2 presents the same dependences at fixed pitch P = 800 nm and NM width *W* varying from 100 to 220 nm. Calculations were performed in accordance with expressions (7) at $\Phi = \varepsilon = 1$, which corresponds to



Figure 1. NM height as a function of effective deposition thickness *vt* at fixed NM width W = 160 nm and various values of pitch *P* (indicated). Formulae (7) with $\Phi = \varepsilon = 1$ were used for calculation.



Figure 2. NM height as a function of effective deposition thickness *vt* at fixed pitch P = 800 nm and various values of NM width *W* (indicated). Formulae (7) with $\Phi = \varepsilon = 1$ were used for calculation.

SAE at elevated supersaturation of the gas medium without desorption of group III element atoms from the NM surface. As was expected, the NM height increases with increasing pitch P and decreasing NM width W. Dependences H(t) become more linear at small P, since the flux of group III element atoms reaches saturation earlier. An intensification of growth at smaller lateral dimensions (in the present case, smaller NM width) is typical of diffusion growth of vertical structures [8–13].

The constructed model allows one to simulate the kinetics of growth and the morphology of NMs in a rectangular geometry that is the most common one for SAE on surfaces with the (111) crystallographic orientation [8]. It was demonstrated that the NM height is a nonlinear function of time and increases with increasing distance between NMs and decreasing NM width. It should be taken into account in growth experiments that the kinetics of growth of NMs on masked substrates and their morphology differ greatly from the corresponding parameters on adsorbing substrates. A separate publication on the intriguing issue of generalization of the obtained results to the case of more complex NM geometries, which are observed in SAE on surfaces with the (100) crystallographic orientation [7], is planned for the future.

Funding

This study was supported by a research grant from the St. Petersburg State University (ID 94033852).

Conflict of interest

The author declares that he has no conflict of interest.

References

- [1] J.A Rogers, M.G. Lagally, R.G. Nuzzo, Nature, 477, 45 (2011). DOI: 10.1038/nature10381
- [2] X. Yuan, D. Pan, Y. Zhou, X. Zhang, K. Peng, B. Zhao, M. Deng, J. He, H.H. Tan, C. Jagadish, Appl. Phys. Rev., 8, 021302 (2021). DOI: 10.1063/5.0044706
- [3] C.-Y. Chi, C.-C. Chang, S. Hu, T.-W. Yeh, S.B. Cronin, P.D. Dapkus, Nano Lett., 13, 2506 (2013).
 DOI: 10.1021/nl400561j
- [4] G. Tutuncuoglu, M. de la Mata, D. Deiana, H. Potts, F. Matteini, J. Arbiol, A. Fontcuberta i Morral, Nanoscale, 7, 19453 (2015). DOI: 10.1039 /C5NR04821D
- [5] P. Aseev, A. Fursina, F. Boekhout, F. Krizek, J.E. Sestoft, F. Borsoi, S. Heedt, G. Wang, L. Binci, S. Martí-Sánchez, T. Swoboda, R. Koops, E. Uccelli, J. Arbiol, P. Krogstrup, L.P. Kouwenhoven, P. Caroff, Nano Lett., **19**, 218 (2019). DOI: 10.1021/acs.nanolett.8b03733
- [6] M. Friedl, K. Cerveny, P. Weigele, G. Tütüncüoglu, S. Marti-Sanchez, C. Huang, T. Patlatiuk, H. Potts, Z. Sun, M.O. Hill, L. Güniat, W. Kim, M. Zamani, V.G. Dubrovskii, J. Arbiol, L.J. Lauhon, D.M. Zumbühl, A. Fontcuberta i Morral, Nano Lett., 18, 2666 (2018). DOI: 10.1021/acs.nanolett.8b00554
- [7] D. Dede, F. Glas, V. Piazza, N. Morgan, M. Friedl, L. Guniat, E. Nur Dayi, A. Balgarkashi, V.G. Dubrovski, A. Fontcuberta i Morral, Nanotechnology, 33, 485604 (2022). DOI: 10.1088/1361-6528/ac88d9
- [8] V.G. Dubrovskii, Phys. Rev. Mater., 7, 026001 (2023).
 DOI: 10.1103/PhysRevMaterials.7.026001
- K. Tomioka, K. Ikejiri, T. Tanaka, J. Motohisa, S. Hara,
 K. Hiruma, T. Fukui, Mater. Res., 26, 2127 (2011).
 DOI: 10.1557/jmr.2011.103
- [10] G.E. Cirlin, V.G. Dubrovskii, N.V. Sibirev, I.P. Soshnikov, Y.B. Samsonenko, A.A. Tonkikh, V.M. Ustinov, Semiconductors, **39**, 557 (2005). DOI: 10.1134/1.1923565.
- [11] L.E Froberg, W. Seifert, J. Johansson, Phys. Rev. B, 76, 153401 (2007). DOI: 10.1103/PhysRevB.76.153401
- [12] M.C. Plante, R.R. LaPierre, J. Appl. Phys., 105, 114304 (2009). DOI: 10.1063/1.3131676

- [13] V.G. Dubrovskii, Yu.Yu. Hervieu, J. Cryst. Growth, 401, 431 (2014). DOI: 10.1016/j.jcrysgro.2014.01.015
- [14] F. Oehler, A. Cattoni, A. Scaccabarozzi, J. Patriarche, F. Glas, J.C. Harmand, Nano Lett., 18, 701 (2018). DOI: 10.1021/acs.nanolett.7b03695
- [15] V.G. Dubrovskii, Nanomaterials, 12, 253 (2022). DOI: 10.3390/nano12020253
- [16] V.G. Dubrovskii, Nanomaterials, 12, 2632 (2022).DOI: 10.3390/nano12152632
- [17] V.G. Dubrovskii, E.D. Leshchenko, Nanomaterials, 12, 1698 (2022). DOI: 10.3390/nano12101698

Translated by D.Safin