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Aspect ratio of III-V ternary nanostructures grown on lattice-mismatched substrates

V.G. Dubrovskii 

St. Petersburg State University, St. Petersburg, Russia

 v.dubrovskii@spbu.ru

ABSTRACT

It is well known that the aspect ratio of heteroepitaxial Stranski-Krastanow or Volmer-Weber islands in different material systems and self-induced GaN nanowires generally increases with the lattice mismatch. An equilibrium model for the composition and preferred aspect ratio of III-V ternary nanostructures $A_xB_{1-x}C$ grown on a lattice-mismatched substrate D is presented. It is shown that the thermodynamically-limited composition is given by the usual equilibrium vapor-solid distribution, while the aspect ratio becomes composition-dependent and increases for nanostructures with larger lattice mismatch. Lower mismatch for modest compositions often leads to two-dimensional growth. The available experimental data for different material systems are analyzed from the viewpoint of the obtained results.

KEYWORDS

nanostructures • elastic relaxation • composition • aspect ratio

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Introduction

Stress-driven Stranski-Krastanow transition from two-dimensional (2D) to three-dimensional (3D) growth in heteroepitaxial systems with modest lattice mismatch (InAs/GaAs [1–4], Ge/Si [4–6] or GaN/AlN [7]), or Volmer-Weber growth of 3D islands on bare substrates with larger lattice mismatch (InAs/Si [8], GaN/Si [9] or GaAs/Al₂O₃ [10]) is widely used for epitaxial synthesis of optoelectronic nanostructures on dissimilar substrates. It is well documented that the aspect ratio of coherent strained islands generally increases with the lattice mismatch, because taller islands are able to relax elastic strain more effectively than flatter ones [2]. In [11], it was noticed that the formation energy of a strained island has a saddle shape when considered as a function of the size and aspect ratio. This provides explicitly the energetically preferred aspect ratio versus lattice mismatch by finding the saddle point coordinates. Vertically aligned III-V nanowires grown by the vapor-liquid-solid method with a metal catalyst [12,13], catalyst-free selective area epitaxy [14] or self-induced mechanism where GaN Volmer-Weber islands on Si(111) substrates subsequently become nanowires [9,15,16] offer often unlimited possibilities for bottom-up design of functional nanomaterials with advanced functionalities. In particular, extremely high aspect ratios of developed III-V nanowires (up to 100) enable dislocation-free growth of nanowire heterostructures in highly mismatched material systems [17] and on lattice-mismatched Si substrates [18].

Compositions of III-V ternary nanostructures [19] and nanowires [20–30] were studied in great detail over the past decades. In particular, the kinetic suppression of wide miscibility gaps in highly mismatched ternary materials was experimentally demonstrated for $In_xGa_{1-x}As$

[20], $\text{In}_x\text{Ga}_{1-x}\text{As}$ [21] and $\text{In}_x\text{Ga}_{1-x}\text{N}$ [22] nanowires. The complex interplay of thermodynamic and kinetic factors influencing the compositions of III-V ternary nanowires is also well understood through the advanced modeling [23–30]. However, these models usually treat 2D III-V ternary monolayers, whereas the influence of a lattice-mismatched substrate from which the nanowire emerges is totally ignored. This is unacceptable for the initial stage of nanowire growth and for self-induced InGaN islands and nanowires on Si substrates. Consequently, in this paper we present the equilibrium modeling of III-V ternary nanostructures of any type (islands, needles, nanowires) $\text{A}_x\text{B}_{1-x}\text{C}$ on lattice-mismatched substrates D that connects the solid and vapor composition with the aspect ratio. The model gives explicitly the 2D or 3D growth domains and the preferred aspect ratio as a function of the nanostructure and vapor composition. These results shed new light on the relationship between the composition and shape of different nanostructures on lattice-mismatched substrates and may be used as a starting point for studies of kinetic aspects of the problem.

Model

The condition for thermodynamic equilibrium for III-V pairs AC and BC in a vapor (or liquid) mother phase and in solid is given by [23,26]:

$$\begin{aligned}\mu_A + \mu_C &= \mu_{AC}, \\ \mu_B + \mu_C &= \mu_{BC}.\end{aligned}\tag{1}$$

Here, μ_k are the chemical potentials of atoms $k = A, B, C$ in a mother phase and μ_{kC} are the chemical potentials of pairs $kC = AC$ and BC in pseudo-binary solid alloy $\text{A}_x\text{B}_{1-x}\text{C}$ with composition x . Here and below, all chemical potentials and other energetic constants are expressed in thermal units. We specifically consider a group III based nanostructure $\text{A}_x\text{B}_{1-x}\text{C}$ (with atoms A and B belonging to group III and atoms C to group V), deposited onto a lattice-mismatched substrate D under high vacuum conditions from vapors of atoms A, B and molecules C_n with pressures P_A, P_B and P_{C_n} ($n = 2$ for dimers As_2, P_2 or N_2 and 4 for tetramers As_4). The total pressure P in a growth chamber is given by $P = P_A + P_B + P_{C_n}$, which is typical for molecular beam epitaxy (MBE). The vapor phase is a mixture of perfect gases with the chemical potentials:

$$\begin{aligned}\mu_A &= \mu_A^P + \ln\left(\frac{P_A}{P}\right) = \mu_A^P + \ln(z\xi), \\ \mu_B &= \mu_B^P + \ln\left(\frac{P_B}{P}\right) = \mu_B^P + \ln[(1-z)\xi], \\ \mu_C &= \frac{1}{n}\left[\mu_{C_n}^P + \ln\left(\frac{P - P_A - P_B}{P}\right)\right] = \mu_C^P + \frac{1}{n}\ln(1-\xi).\end{aligned}\tag{2}$$

Here, μ_A^P, μ_B^P and $\mu_C^P = \mu_{C_n}^P/n$ are the atomic chemical potentials of the three species in vapor at the total pressure $P = \text{const}$. The quantities

$$\begin{aligned}z &= \frac{P_A}{P_A + P_B}, \\ \xi &= \frac{P_A + P_B}{P},\end{aligned}\tag{3}$$

denote the vapor composition and the dimensionless partial pressure of group III atoms A and B, respectively. In the ternary liquid ABC with atomic concentrations χ_A, χ_B and $\chi_C = 1 - \chi_A - \chi_B$ which is a mother phase for self-catalyzed vapor-liquid-solid growth of nanowires [23–26], one has $n = 1$, with $z = y = \chi_A/(\chi_A + \chi_B)$ as the liquid composition and $\xi = \chi_A + \chi_B = \chi_{\text{tot}}$ as the total concentration of group III atoms in liquid.

Our model for the chemical potentials of pairs AC and BC in solid is given by:

$$\begin{aligned}\mu_{AC} &= \mu_{AC}^P + \ln x + \omega(1-x)^2 - u(x, \beta), \\ \mu_{BC} &= \mu_{BC}^P + \ln(1-x) + \omega x^2 - u(x, \beta).\end{aligned}\tag{4}$$

The first three terms in these expressions correspond to the regular solution model for pseudo-binary alloys [13,23–26], with ω as the pseudo-binary interaction parameter in thermal units. The chemical potentials μ_{AC}^P and μ_{BC}^P correspond to pure binary solids AC and BC. Thermodynamically forbidden miscibility gap is present whenever $\omega > 2$ at a growth temperature. For example, the miscibility gap closed at 543 °C for $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy [24] and at ~ 1250 °C for $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloy [22]. The last term u describes the influence of elastic stress induced by lattice mismatch when a nanostructure $\text{A}_x\text{B}_{1-x}\text{C}$ with the aspect ratio β (the ratio of nanostructure height over length of its base) is grown on a dissimilar substrate D. This contribution is the same for equally strained pairs AC and BC. Using the results of Refs. [2,7,11], the function $u(x, \beta)$ for ternary nanostructures with composition x can be put in the form:

$$u(x, \beta) = \lambda(x)\varepsilon^2(x)\varphi(\beta) \cong \frac{\lambda\varepsilon^2(x)}{1+\beta/\beta_*}. \quad (5)$$

Here, $\lambda(x) = E(x)\Omega/(1 - \nu)$ is the elastic energy per “pair” $\text{A}_x\text{B}_{1-x}\text{C}$ in a uniformly strained 2D layer, with $E(x)$ as the Young’s modulus of ternary material, ν as the Poisson ratio ($\nu = 1/3$ for cubic zincblende materials), Ω as the elementary volume per III-V pair in solid, and $\varepsilon(x)$ as the composition-dependent lattice mismatch parameter. The function $\varphi(\beta)$ measures the effect of strain relaxation at free sidewalls of a nanostructure [2,7,11,17]. Although $\varphi(\beta)$ generally depends on the nanostructure geometry (cylinder, cone, truncated cone or pyramid etc.), it can often be approximated by the simplest expression $1 + \beta/\beta_*$, with a small $\beta_* \sim 0.1$ [2]. The smallness of the characteristic β_* shows that the elastic relaxation on free sidewalls of 3D island increases sharply with its aspect ratio. Taking the simplest linear approximation for the Young’s modulus and lattice constant between the two binaries, one arrives at:

$$u(x, \beta) = \frac{[\lambda_{AC}x + \lambda_{BC}(1-x)][\varepsilon_{AC}x + \varepsilon_{BC}(1-x)]^2}{1+\beta/\beta_*}. \quad (6)$$

Substitution of Eqs. (2), (4) and (6) into Eqs. (1) yields two equations for the unknowns x and β that contain the technologically controlled parameters of the vapor composition z and ξ and chemical parameters of a ternary material (chemical potentials of pure phases and pseudo-binary interaction parameter). In the next section, we analyze the corresponding solutions under thermodynamic equilibrium and their predictions for the composition and aspect ratio of III-V ternary nanostructures on lattice-mismatched substrates.

Results and Discussion

Taking the difference of Eqs. (1), using Eqs. (2) for μ_A, μ_B and Eqs. (4) for μ_{AC}, μ_{BC} , we obtain the equilibrium vapor-solid distribution [23,26] that connects the composition of vapor and solid:

$$\begin{aligned} Z &= \frac{1}{1 + \alpha[x/(1-x)]e^{\omega(2x-1)}} \\ \alpha &= e^{\Delta\mu_{AC}^P - \Delta\mu_{BC}^P}. \end{aligned} \quad (7)$$

This distribution is fully determined by the pseudo-binary interaction parameter ω and the affinity parameter α , with $\Delta\mu_{AC}^P = \mu_A^P + \mu_C^P - \mu_{AC}^P$, $\Delta\mu_{BC}^P = \mu_B^P + \mu_C^P - \mu_{BC}^P$ as the well-defined differences of the chemical potentials for pairs AC and BC in pure vapor and binary solid phases. The equilibrium vapor-solid distribution is independent of the elastic energy u that cancels in the difference of Eqs. (4). Equation (7) is equivalent to the nucleation-limited distribution at a composition-independent surface energy of III-V ternary island [24]. The nucleation-limited composition is obtained by minimizing the saddle point of the formation energy as a function of the island size and composition [24,26] Figure 1 shows the vapor-solid distribution given by Eq. (7) for $\text{In}_x\text{Ga}_{1-x}\text{As}$ material at 450 and 570 °C. These temperatures

correspond to the presence ($\omega = 2.373$) and absence ($\omega = 1.906$) of the miscibility gap at lower and higher temperature, respectively. The low affinity of In atoms with respect to Ga ($\alpha \cong 0.033$ at 470 °C and 0.05 at 570 °C) requires high (more than 0.8) In fractions in a mother phase for obtaining any reasonable InAs fraction in solid in both cases [25–27].

Let us now analyze the relationship between the island aspect ratio β , solid composition x and partial pressure of group III atoms in vapor ξ . At small lattice mismatch corresponding to $u \rightarrow 0$ in Eqs. (4), any of these two equations yield the result:

$$\xi_0(1 - \xi_0)^{1/n} = e^{-\Delta\mu_{AC}^P x_0} e^{\omega(1-x_0)^2} + e^{-\Delta\mu_{BC}^P(1-x_0)} e^{\omega x_0^2}, \quad (8)$$

with $\beta \rightarrow 0$. At $n = 1$ and $\xi = \chi_A + \chi_B = \chi_{tot}$, this is reduced to the known result for the equilibrium concentration of atoms A and B in liquid catalyst droplets on top of vapor-liquid-solid nanowires that grow in 2D mode [23]. At larger lattice mismatch, Eqs. (4) predict the 2D-to-3D growth transition, with the preferred aspect ratio given by:

$$\frac{\beta}{\beta_*} = \frac{[\lambda_{AC}x + \lambda_{BC}(1-x)][\varepsilon_{AC}x + \varepsilon_{BC}(1-x)]^2}{F(x, \xi)} - 1. \quad (9)$$

Here, the function $F(x, \xi)$ depends on the composition of 3D island and partial pressure of group III vapors as follows:

$$F(x, \xi) = -\Delta\mu_{AC}^P - \ln[\xi(1 - \xi)^{1/n}] + \ln x + \omega(1 - x)^2 + \ln \left[1 + \alpha \frac{x}{1-x} e^{\omega(2x-1)} \right]. \quad (10)$$

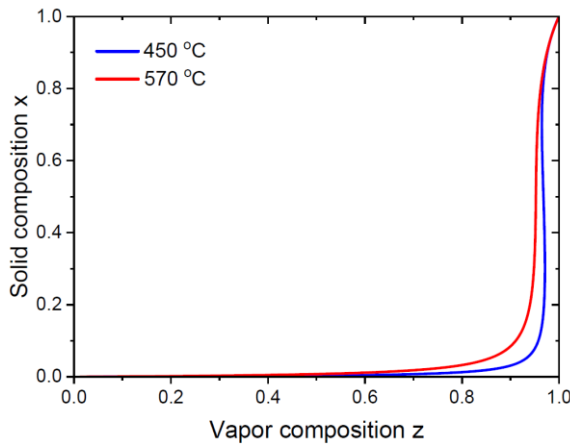


Fig. 1. Vapor-solid distributions of $\text{In}_x\text{Ga}_{1-x}\text{As}$ nanostructures at two different temperatures, obtained from Eq. (7) with $\omega = 2.373$, $\alpha = 0.033$ at 450 °C and $\omega = 1.906$, $\alpha = 0.05$ at 570 °C

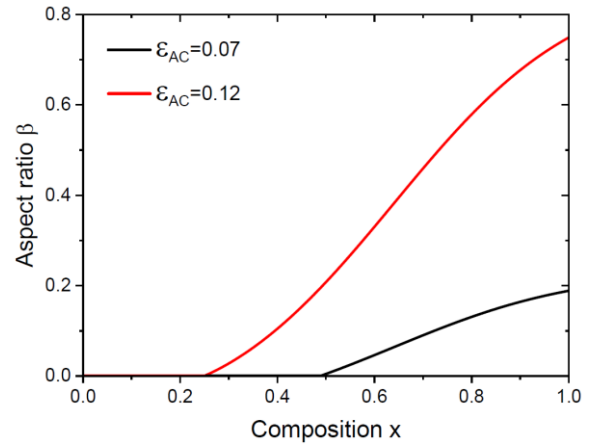


Fig. 2. Aspect ratio of 3D islands $\text{A}_x\text{B}_{1-x}\text{C}/\text{BC}$ versus their composition x at $\varepsilon_{AC} = 0.07$ and 0.12. The curves are obtained from Eq. (12) with $\beta_* = 0.1$, $\lambda_{AC} = 375$, fixed $\alpha = 0.021$ and $\xi_0(1 - \xi_0)^{1/n} / \xi(1 - \xi)^{1/n} = 0.05$

Any negative solutions given by Eq. (9) correspond to planar 2D growth at $\beta \rightarrow 0$. For binary islands AC/D, Eqs. (9) and (10) at $x = 1$ give:

$$\frac{\beta}{\beta_*} = \frac{\lambda_{AC}\varepsilon_{AC}^2}{-\Delta\mu_{AC}^P - \ln[\xi(1 - \xi)^{1/n}]} - 1. \quad (11)$$

According to this expression, the equilibrium aspect ratio scales with $\lambda_{AC}\varepsilon_{AC}^2$ and is proportional to the effective supersaturation of vapor (the denominator in Eq. (11) should be positive for 3D islands). This result of our equilibrium model is consistent with [11], where the preferred aspect ratio was obtained in the nucleation-limited regime. Considering heteroepitaxial growth of ternary material $\text{A}_x\text{B}_{1-x}\text{C}$ on BC with zero lattice mismatch $\varepsilon_{BC} = 0$, and assuming equilibrium of 2D layer BC with vapor at pressure ξ_0 , from Eq. (8) one has $\xi_0(1 - \xi_0)^{1/n} = \exp(-\Delta\mu_{BC}^P)$. This reduces Eqs. (9) and (10) to:

$$\frac{\beta}{\beta_*} = \frac{\lambda_{AC}\varepsilon_{AC}^2 x^2}{\ln \left[\frac{\xi_0(1 - \xi_0)^{1/n}}{\xi(1 - \xi)^{1/n}} \right] - \alpha + \ln x + \omega(1 - x)^2 + \ln \left[1 + \alpha \frac{x}{1-x} e^{\omega(2x-1)} \right]} - 1, \quad (12)$$

for any $A_xB_{1-x}C/BC$ material system. Again, the transition from 2D to 3D growth occurs only for positive β , while smaller lattice misfits correspond to 2D growth of uniformly strained wetting layer [2,6]. Table 1 summarizes the values of $E/(1-\nu)$ and the experimentally measured aspect ratios β_{exp} for some interesting heteroepitaxial systems in a wide range of ε_{AC} from 0.04 to 0.22 [4–10]. With a typical value of $E/(1-\nu) \sim 100$ GPa and $\Omega = 0.0452$ nm³ for zincblende GaAs, we get a typical value of $\lambda_{AC} \cong 28.2$ eV. This corresponds to a dimensionless elastic modulus of 375 at 600 °C, $\lambda_{AC}\varepsilon_{AC}^2 \cong 1.86$ at $\varepsilon_{AC} = 0.07$ and 5.47 at $\varepsilon_{AC} = 0.12$. Figure 2 shows the equilibrium aspect ratios versus composition for these lattice mismatch parameters, obtained from Eq. (12) at a fixed $\beta_* = 0.1$, $\alpha = 0.021$ and $\xi_0(1-\xi_0)^{1/n}/\xi(1-\xi)^{1/n} = 0.05$. One can see that increasing the lattice mismatch in AC binary material from 0.07 to 0.12 increases the largest aspect ratio at $x = 1$ from ~ 0.19 to ~ 0.75 , and extends the 3D growth domain from $x > 0.25$ to $x > 0.5$ for these parameters.

Table 1. Parameters of different binary lattice-mismatched systems

Material system	$E/(1-\nu)$, GPa	ε_{AC}	β_{exp}	Ref.
Ge/Si	139	0.04	0.1-0.33	[4]
GaN/AlN	279	0.04	0.31	[5]
InAs/GaAs	79	0.07	0.23-0.71	[4,6,7]
InAs/Si	79	0.12	0.65-0.95	[8]
GaN/Si	279	0.17	2-3	[9]
ZnO/Al ₂ O ₃	209	0.22	3-5	[10]

In MBE growth under group V rich conditions in vapor, one can assume that $\xi \ll 1$, because the total pressure of group III atoms is usually much less than the pressure of group V species (As, P or N molecules). The pressure difference is largely enhanced by re-emitted group V species [13]. In this case, one can use the approximation $\xi(1-\xi)^{1/n} \cong \xi$ in the above expressions. Figure 3 shows how the island aspect ratio increases with the total pressure of group III atoms at a fixed $\lambda_{AC}\varepsilon_{AC}^2 = 5.47$. The curves are obtained from Eq. (12) with the same parameters as in Fig. 3, a fixed $\xi_0 = 0.01$ and varying pressures ξ . According to the figure, formation of 3D ternary islands is strongly suppressed at low pressures of group III atoms (or strongly group V rich growth conditions) and enhanced otherwise. Finally, Fig. 4 shows the data on the experimentally observed aspect ratios for different material systems from Table, fitted by the approximate expression $\beta = \beta_*(856\varepsilon_{AC}^2 - 1)$. Of course, this oversimplified Eq. (11) gives only the qualitative trend agreement, because binary supersaturations for each growth are not exactly known and the elastic constants of different binary materials vary quite significantly according to Table 1. Furthermore, our equilibrium model does not account for the kinetic effects such as the Stranski-Krastanow growth transition in Ge/Si and InAs/GaAs systems upon reaching the critical thickness of metastable wetting layer [2,4,6]. However, the general trend for the quadratic increase of the aspect ratio of 3D islands with the lattice mismatch ε_{AC}^2 is well reproduced by the model.

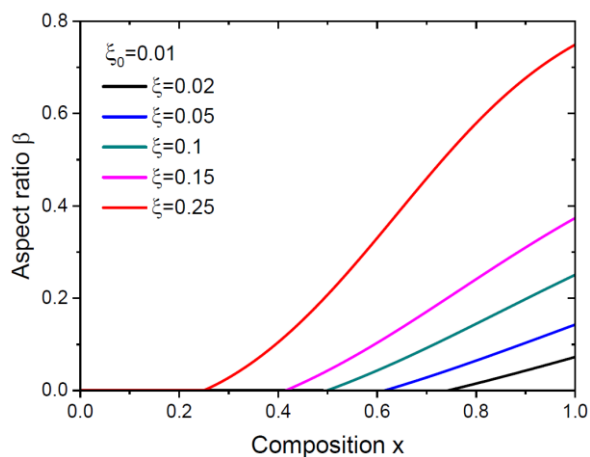


Fig. 3. Extension of 3D growth domain and increase of the aspect ratios of ternary islands $A_xB_{1-x}C/BC$ with partial pressure of group III atoms ξ

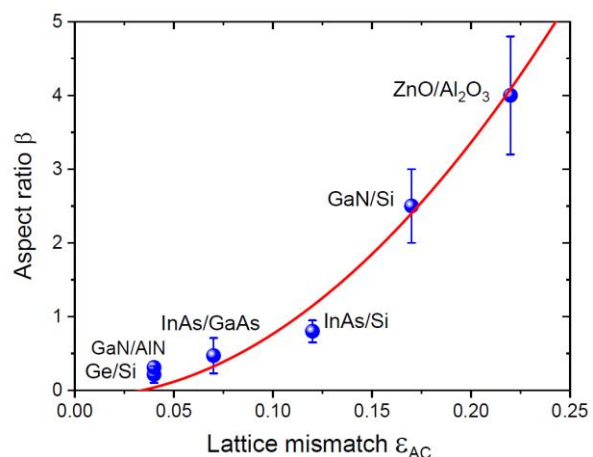


Fig. 4. Quadratic increase of the aspect ratio of binary islands in different heteroepitaxial systems from Table 1 with the lattice mismatch: experimental data (symbols) and model fit (line)

Conclusions

In summary, the developed equilibrium model provides explicitly the aspect ratio of III-V ternary islands or nanowires as a function of their composition and partial pressure of group III atoms. For any ternary material $A_xB_{1-x}C$ on a lattice-mismatched substrate D with larger lattice mismatch for higher composition x , the aspect ratio increases with x . This is valid also in the particular case of $A_xB_{1-x}C/BC$ system. Importantly, the critical composition for 2D-to-3D growth transition and the aspect ratio of islands in 3D growth domain sharply increase with pressure of group III atoms. Highly group V rich growth conditions suppress 3D islanding. The vapor-solid distribution is not affected by the elastic stress relaxation and remains the same for 2D layers and 3D islands. We hope that these results will be useful for general understanding, modeling and planning the growth experiments for obtaining the required shapes and compositions in a wide range of III-V material systems and on different dissimilar substrates including Si. The presented approach requires a refinement to account for the kinetic growth effects. We plan to consider this interesting problem in a future paper.

CRedit authorship contribution statement

Vladimir G. Dubrovskii  **Sc**: writing – review & editing, writing – original draft; conceptualization.

Conflict of interest

The author declares that he has no conflict of interest.

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