Thermodynamic analysis of semipolar GaN and AlN under metalorganic vapor phase epitaxy growth conditions

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We perform an improved method of thermodynamic analysis for semipolar (1122) and (1101) surfaces of GaN and AlN to elucidate the relationship between growth conditions and semipolar and polar surfaces during metalorganic vapor phase epitaxy (MOVPE). The calculations for H₂ carrier gas suggest that for both GaN and AlN the maximum temperature for growth on (1122) surfaces is higher than that for growth on (0001) surfaces. On the other hand, the maximum temperature for growth on GaN (100) surfaces is comparable to that for growth on AlN (0001) surfaces, while the maximum temperature for the growth on AlN (0001) surfaces is higher than that for growth on AlN (100) surfaces. These results could be used to provide favorable conditions for growth of group-III nitrides along a semipolar orientation during MOVPE.

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1. Introduction

Group-III nitrides including GaN and AlN have attracted much attention for applications in optoelectronic devices such as laser diodes and light-emitting diodes.¹⁻⁸ In order to fabricate these devices, the growth of high-quality layers has been carried out along various orientations using epitaxial growth methods such as metalorganic vapor phase epitaxy (MOVPE), hydride vapor phase epitaxy (HVPE) and molecular beam epitaxy (MBE). In general, epitaxial growth of Group-III nitrides has been performed along the polar [0001] direction.⁹⁻¹⁴ However, it is well known that [0001]-oriented group-III nitrides exhibit a strong electronic field produced by spontaneous and piezoelectric fields, which causes reduced radiative efficiency and redshift of optical transitions. To overcome these problems, there has been increasing interest in epitaxial growth along nonpolar and semipolar orientations.¹⁵⁻¹⁷ Despite these experimental studies, understanding of the optimum growth conditions and their dependence on orientation is still lacking. This could be elucidated by understanding the growth reactions on a reconstructed semipolar surface, which depend on growth conditions such as partial pressure and temperature.

In our previous studies we proposed an approach for calculating the absolute surface energy of semipolar planes such as (1122) and (1101) using a wedge-shape geometry method.¹⁸,¹⁹ This approach enabled us to directly compare the absolute surface energy in a semipolar orientation with that in a polar orientation. Therefore, our calculated results revealed the relative stability between semipolar and polar surfaces. However, these absolute surface energies do not incorporate the experimental conditions such as the temperature and partial pressure of molecules supplied during epitaxial growth. To compare the theoretical results with experimental results, we recently proposed an improved thermodynamic analysis that incorporates absolute surface energies on GaN of polar and nonpolar planes obtained by ab initio calculations.²⁰,²¹ According to a thermodynamic study, growth on GaN (0001) surfaces is possible at higher temperatures than on (0001) surfaces during MOVPE.²² As a result, the maximum growth temperature on polar surfaces determined by the thermodynamic approach enables us to compare the calculated results with experiments directly. However, the maximum temperature of semipolar group-III nitride surfaces under MOVPE growth conditions is still unclear. To discuss the relationship of growth conditions among various surface orientations, thermodynamic analysis of group-III nitrides on semipolar surfaces is required.

In this study, we perform the improved thermodynamic approach for semipolar (1122) and (1101) surfaces of Group-III nitrides. The analysis is carried out for both semipolar and polar AlN surfaces, because the maximum temperature of MOVPE growth on polar AlN surfaces has not yet been clarified. In this paper, the stability of polar and semipolar surfaces such as (1122) and (1101) taking account of hydrogen atoms is determined using absolute surface energies. Furthermore, the orientation dependence of growth conditions is discussed on the basis of the temperature dependence of equilibrium partial pressure. Our calculations demonstrate characteristic features of the growth reaction on semipolar surfaces under MOVPE growth conditions. Section 2 briefly explains the computational method, presenting ab initio calculations and thermodynamic analysis. The results and discussion for semipolar GaN surfaces, polar AlN surfaces and semipolar AlN surfaces are given out in Sects. 3.1–3.3, respectively. In Sect. 3.4, the differences in growth conditions, such as maximum temperature and carrier gases, are also discussed. Finally, we summarize this study in Sect. 4.

2. Computational methods

The total energy calculations are performed within the plane-wave pseudopotential approach using the generalized gradient approximation.²² We use norm-conserving pseudopotentials for Al, Ga and H atoms²³ and ultrasoft pseudopotentials for N
Atoms. Ga 3d electrons are treated by partial core corrections. The conjugate-gradient technique is utilized for both the electronic structure calculations and geometric optimization. The valence function expansions are expanded by the plane-wave basis set with a cut-off energy of 30.25 Ry. We use 72 k-point sampling for the 1 × 1 surface unit, which provides sufficient accuracy for the total energy. The computations are performed using the Tokyo Ab initio Program Package.

The semipolar surfaces are simulated using 1 × 1, 2 × 2, c(2 × 2) and 1 × 2 slab models consisting of seven bilayers and a vacuum region of approximately 10 Å. The bottom surface of the slab is passivated with fractional hydrogen atoms and the lowermost four layers are fixed at ideal positions. These atomic configurations are considered according to previous calculations and the electron counting (EC) rule. The relationships between them are expressed as μ_Ga = μ_Ga^bulk and μ_AL = μ_AL^bulk = μ_AL^bulk where μ_Ga^bulk and μ_AL^bulk are the chemical potentials of Ga and Al, respectively, and μ_GaN and μ_ALN are total energies of bulk GaN and AlN, respectively. The absolute surface energies are estimated as a function of μ_Ga or μ_AL, which can vary in the thermodynamically allowed range. This range corresponds to the enthalpy of the heat of formation, and the calculated values for GaN and AlN are −1.2 eV and −3.0 eV, respectively.

Therefore, the ranges of μ_Ga and μ_AL are −1.2 eV ≤ μ_Ga ≤ μ_Ga^bulk and μ_AL^bulk −3.0 eV ≤ μ_AL ≤ μ_AL^bulk where μ_Ga^bulk and μ_AL^bulk are the chemical potentials of bulk Ga and Al, respectively.

To analyze the temperature dependence of equilibrium partial pressure on semipolar surfaces it is necessary to determine what kinds of surface reconstruction appear on growth surfaces. The surface reconstruction with the lowest absolute surface energy is the reconstructed and ideal surface, respectively. The absolute surface energies are estimated as a function of μ_Ga or μ_AL, which can vary in the thermodynamically allowed range. This range corresponds to the enthalpy of the heat of formation, and the calculated values for GaN and AlN are −1.2 eV and −3.0 eV, respectively.

Therefore, the ranges of μ_Ga and μ_AL are −1.2 eV ≤ μ_Ga ≤ μ_Ga^bulk and μ_AL^bulk −3.0 eV ≤ μ_AL ≤ μ_AL^bulk where μ_Ga^bulk and μ_AL^bulk are the chemical potentials of bulk Ga and Al, respectively.

According to the second law of thermodynamics, the equilibrium condition of this reaction is shown as

$$\Delta G_{\text{surface-gas}}^0 + RT \ln \left[ \frac{n_{\text{GaN}}(P_{\text{H}_2})^{3/2}}{P_{\text{Ga}}P_{\text{NH}_3}} \right] = 0,$$

where \(\Delta G_{\text{surface-gas}}^0\) is the standard Gibbs energy of the reaction in Eq. (5). R is the gas constant and \(n_{\text{GaN}}\) is the activity of GaN that defines a value of 1. The assumption of a stoichiometric growth conditions is given by

$$\Delta p_{\text{Ga}} = \Delta p_{\text{NH}_3},$$

$$\Delta p_{\text{Ga}} = -\frac{2}{3} \Delta p_{\text{H}_2},$$

where \(\Delta p\) shows the difference between the decomposed partial pressure \(p'\) and \(p\). The gas-surface equilibrium partial pressures \(P_{\text{Ga}}, P_{\text{NH}_3}\), and \(P_{\text{H}_2}\) are obtained by solving Eq. (6) under the stoichiometric growth conditions of Eqs. (7) and (8).

The standard Gibbs free energy \(\Delta G_{\text{surface-gas}}^0\) is described as

$$\Delta G_{\text{surface-gas}}^0 = \Delta G_{\text{bulk-gas}}^0 + \Delta E_{\text{surface-bulk}}^0,$$

where \(\Delta G_{\text{bulk-gas}}^0\) is the standard Gibbs free energy of the reaction from source gas to solid bulk. The energy difference between the surface and bulk phase \(\Delta E_{\text{surface-bulk}}\) is given by

$$\Delta E_{\text{surface-bulk}} = \{ E_{\text{slab}} - \mu_{\text{Ga}} n_{\text{Ga}} - \mu_{\text{N}} n_{\text{N}} - \mu_{\text{H}} n_{\text{H}} - A_{\text{slab}} \sigma_{\text{pass}} \}/ (n_{\text{Ga}}^\text{top} + n_{\text{Ga}}^\text{ad} + n_{\text{N}}^\text{ad}).$$

Here, \(E_{\text{slab}}\) is total energy of the slab model. \(n_{\text{Ga}}, n_{\text{N}}\), and \(n_{\text{H}}\) are the number of atoms of Ga, N, and H, respectively, in the slab model. As before, \(\mu_{\text{Ga}}, \mu_{\text{N}}, \mu_{\text{H}}\) are the chemical potentials of Ga, N, and H, respectively, \(\sigma_{\text{pass}}\) is the semipolar surface energy of the bottom surface which is passivated with fractional H atoms. The approach to calculate \(\sigma_{\text{pass}}\) is based on the use of a wedge-shaped geometry. Details of the
calculation procedure are described elsewhere.\(^{18,19}\) \(n\) is the number of GaN formula units in the topmost layers, respectively. \(A_{\text{slab}}\) is the surface area of the slab model.

3. Results and discussion

3.1. Semipolar GaN surfaces

Figure 1 shows the surface phase diagrams of GaN(1\(\overline{1}22\)) and (1\(\overline{1}01\)) for \(N_2\) and \(H_2\) carrier gases as functions of temperature and N/III ratio, where III denotes group-III elements such as Ga and Al. Details of the growth conditions are given in the figure. For GaN(1\(\overline{1}22\)) surfaces, the surface reconstruction terminated with hydrogen atoms (3N–H+NH\(_2\)) appears over the entire range of temperature for both \(N_2\) and \(H_2\) carrier gases, as shown in Figs. 1(a) and 1(b). This is because the Gibbs free energy in Eq. (1) for 3N–H+NH\(_2\) is much lower than that for other GaN(1\(\overline{1}22\)) surface reconstructions. Furthermore, for GaN(1\(\overline{1}01\)) surfaces, a structural change from the surface reconstruction covered by Ga atoms (Ga monolayer) to that with hydrogen atoms (2NH\(_2\)+5N–H) is seen in the case of both \(N_2\) and \(H_2\) carrier gases. However, the phase transition temperature is different depending on the carrier gas. For \(N_2\) carrier gas, 2NH\(_2\)+5N–H appears at a lower temperature than 995 °C with a V/III ratio (V denotes group-V element) of 2000 in Fig. 1(c), while in the case of \(H_2\) carrier gas, the phase transition temperature is found to be 900 °C at a V/III ratio of 2000 [Fig. 1(d)].

The temperature dependence of the equilibrium partial pressure of Ga, \(p_{\text{Ga}}\), with \(N_2\) and \(H_2\) carrier gases on semipolar GaN surfaces is shown in Fig. 2. In the case of GaN(1\(\overline{1}22\)) growth under \(N_2\) carrier gas in Fig. 2(a), the driving force \(\Delta p_{\text{Ga}}\) (\(\Delta p_{\text{Ga}} = p_{\text{Ga}}^0 - p_{\text{Ga}}\), where \(p_{\text{Ga}}^0\) is the input Ga partial pressure) for GaN(1\(\overline{1}22\)) surfaces is always positive over a wide range of temperature, similar to the case of the GaN(0001) surface.\(^{21}\) Moreover, \(\Delta p_{\text{Ga}}\) on GaN(1\(\overline{1}01\)) and (0001) surfaces becomes negative when the temperature is higher than approximately 1105 °C [Fig. 2(c)] and 1075 °C,\(^{21}\) respectively. It is thus expected that growth on GaN(1\(\overline{1}22\)) surfaces is possible at a higher temperature than on GaN(0001) surfaces, while the maximum growth temperature on GaN(1\(\overline{1}01\)) surfaces is similar to that on GaN(0001) surfaces. It should be noted that the maximum temperature for growth on GaN(0001) surfaces is found to be similar to
that for growth on GaN(1122) surfaces. However, growth on GaN(0001) surfaces is possible at a higher temperature than growth on GaN(1101) surfaces.

### 3.2. Polar AlN surfaces

Figure 3 shows surface phase diagrams for polar AlN with N2 and H2 carrier gases as functions of temperature and V/III ratio. For N2 carrier gas, the surface reconstruction terminated by NH3 and NH2 (Al–NH3 + 3Al–NH2) is found at low temperature regardless of the V/III ratio, as shown in Fig. 3(a). Since the H atoms desorb from the surface with increasing temperature, reconstructed surfaces covered by N atoms (N ad–H + Al–NH2 and N adatom) are stabilized [Fig. 3(a)]. On the other hand, for H2 carrier gas the surface reconstruction with NH3 and NH2 (Al–NH3 + 3Al–NH2) on AlN(0001) surfaces is seen even at high temperatures over a wide range of V/III ratios, as shown in Fig. 3(b). For AlN (0001) surfaces, the surface reconstructions covered by Al atoms (Al adlayer and Al adatom) appear even at high temperature over a wide range of V/III ratios for N2 carrier gas [Fig. 3(c)]. A structural change from surface reconstruction with an Al layer (Al adlayer) to that with hydrogen (3N–H) is recognized under H2 carrier gas, as shown in Fig. 3(d).

The temperature dependence of the equilibrium partial pressure of Al, \( p_{Al} \), with N2 and H2 carrier gases on polar AlN surfaces is shown in Fig. 4. The figure implies that the driving force \( \Delta p_{Al} := p_{Al}^0 - p_{Al} \) (where \( p_{Al}^0 \) is the input Al partial pressure) of polar AlN surfaces is always positive over a wide range of temperature for N2 carrier gas [see Figs. 4(a) and 4(c)]. Therefore, the maximum growth temperature on AlN(0001) surfaces is comparable to that on AlN(0001).
surfaces. In contrast, under H2 carrier gas, growth on polar AlN(0001) and (001) is inhibited around 1530 °C and 1770 °C, as shown in Figs. 4(b) and 4(d), respectively. As a result, growth is possible on AlN(0001) surfaces at higher temperatures than on AlN(0001) surfaces.

3.3. Semipolar AlN surfaces

Figure 5 shows surface phase diagrams of AlN(1122) and (1101) for N2 and H2 carrier gases as functions of temperature and V/III ratio. Surface reconstruction terminated by hydrogen (2NH2 + 14N–H) on AlN(1122) is found at low temperatures, and reconstructions covered by Al and N \{c(2 × 2) Al_{ad} + N_{ad}\} appear at high temperature over a wide range of V/III ratios for both N2 and H2 carrier gases [see Figs. 5(a) and 5(b)]. However, the phase transition temperature differs depending on the carrier gas. For N2 carrier gas, the phase transition temperature is found to be 1410 °C, while for H2 carrier gas the Al_{ad} + N_{ad} surface reconstruction appears at high temperatures above 1630 °C. Furthermore, for AlN(1101) under N2 carrier gas, reconstructed surfaces covered by Al atoms (Al bilayer and Al monolayer) are stable surface reconstructions over a wide temperature range, as shown in Fig. 5(c). On the other hand, for an AlN(1101) surface with H2 carrier gas, structural variation from surface reconstruction with Al layers (Al bilayer and Al monolayer) to one with hydrogen (2NH2 + 5N–H) is seen around 1360 °C with a V/III ratio of 1000, then the N desorbed surface reconstruction (3N_{ad}) appears at an extremely high temperature [1740 °C in Fig. 5(d)].

The relationship between temperature and \(p_{Al}\) for N2 and H2 carrier gases on semipolar AlN surfaces is shown in Fig. 6. For N2 carrier gas, the \(\Delta p_{Al}\) of the AlN(1122) surface always becomes positive even at high temperature [Fig. 6(a)], whereas growth on AlN(1101) is interrupted at around 1650 °C with a N/III ratio of 1000 [Fig. 6(c)]. Moreover, growth on semipolar AlN(1122) and (1101) surfaces for H2 carrier gas with a V/III ratio of 1000 is suppressed around 1790 °C and 1480 °C, respectively, as shown Figs. 6(b) and 6(d). As a result, it is expected that for N2 carrier gas the maximum growth temperature on an AlN(1122) surface is comparable to that on an AlN(0001) surface, while for H2 carrier gas growth on AlN(1122) surfaces can occur at a higher temperature than on AlN(0001) surfaces. Furthermore, the maximum temperature for growth on AlN(1122) surfaces is similar to that for AlN(0001) surfaces regardless of the carrier gas. Growth on polar AlN surfaces is possible at higher temperatures than growth on AlN(1101) surfaces for both N2 and H2 carrier gases.

3.4. Difference in growth conditions between GaN and AlN surfaces

Table I shows the difference in growth conditions between GaN and AlN surfaces. The major features of the growth conditions are discussed in the following.
3.4.1. Comparison of growth conditions on semipolar surfaces with those on (0001) surfaces. Growth on (1122) surfaces is possible at a higher temperature than on (0001) surfaces for both GaN and AlN for H2 carrier gas. In contrast, the maximum temperature for growth on AlN(1122) surfaces for N2 carrier gas is similar to that for growth on AlN(0001) surfaces. This can be understood as follows: the difference in the maximum growth temperature between N2 and H2 carrier gases in AlN can be explained in terms of the difference in surface reconstruction depending on the carrier gas, as shown in Figs. 3(a) and 3(b). This difference in the reconstructed surface leads to a difference in \( \Delta E_{\text{surface-bulk}} \) in Eq. (9) depending on the carrier gas. According to Eq. (9), \( \Delta E_{\text{surface-gas}} \) becomes small for small \( \Delta E_{\text{surface-bulk}} \), resulting in small \( \Delta p_{\text{H2}} \) in Eq. (8). This corresponds to a large \( \Delta p_{\text{Ga}} \), leading to high maximum growth temperature. Indeed, the calculated \( \Delta E_{\text{surface-bulk}} \) on an AlN(0001) surface for N2 carrier gas is as small as that on an AlN(1122) surface, while \( \Delta E_{\text{surface-bulk}} \) on an AlN(0001) surface for H2 carrier gas is quite large.

The maximum growth temperature for (1T01) and (0001) surfaces depends on the group-III element. In the case of GaN, the maximum temperature for growth on a GaN(1T01) surface is comparable to that on a GaN(0001) surface. On the other hand, for AlN, growth on an AlN(0001) surface is possible at higher temperatures than for growth on an AlN (1T01) surface. The higher temperature for growth on an AlN (0001) surface than for an AlN(1T01) surface originates from the presence of Nde, as shown in Figs. 5(c) and 5(d). Owing to the large \( \Delta E_{\text{surface-bulk}} \) in Eq. (9) and the large surface
Fig. 5. (Color online) Surface phase diagrams for AlN(1122) and (1101) for both N2 and H2 carrier gases. F is the ratio of H2 in the carrier gas. Growth conditions are as follows: $p_{\text{tot}} = 1$ atm, $p_{\text{NH3}} = 0.05$ atm, $\alpha = 0.25$, $F = 0$ or 1, where $p_{\text{tot}}$ is total pressure, $p_{\text{NH3}}$ is input ammonia pressure and $\alpha$ is the decomposition ratio of ammonia.

Fig. 6. (Color online) Gas-surface equilibrium Al partial pressure, $p_{\text{Al}}$, under MOVPE growth conditions as a function of temperature for AlN(1122) and (1101) surfaces. Black, green, red and blue lines represent the equilibrium Al partial pressure of the bulk phase, input partial pressure $p_{\text{Al}}^0$ and the lower and upper limit of $p_{\text{Al}}$, respectively. When $p_{\text{Al}}$ is lower than $p_{\text{Al}}^0$, growth proceeds smoothly. On the other hand, growth is inhibited when $p_{\text{Al}}$ is higher than $p_{\text{Al}}^0$. © 2019 The Japan Society of Applied Physics.
energy of $N_{\text{de}}$, the driving force on an AlN(1101) surface is smaller than that on an AlN(0001) surface.

3.4.2. Comparison of growth conditions on semipolar surfaces with those on (0001) surfaces. For GaN surfaces, the maximum growth temperature depends on the carrier gas. For $N_2$ carrier gas, the maximum temperature for growth on a GaN(1122) surface is comparable to that for growth on a GaN(0001) surface. In contrast, for $H_2$ carrier gas, the maximum temperature for growth on a GaN(1122) surface is always higher than that for a GaN(1122) surface.

On the other hand, the maximum growth temperature on AlN(1122) surfaces is similar to that on AlN(0001) surfaces regardless of the carrier gas. These trends are explained by considering the equilibrium constant in Eq. (5) and the relationship between $\Delta P_{N_2}$ and $\Delta P_{H_2}$ in Eq. (8), as mentioned in Sect. 3.4.1. Growth on (0001) surfaces is possible at higher temperatures than growth on a (1101) surface regardless of the group-III element and carrier gas. This is because $\Delta E_{\text{surface-bulk}}$ in Eq. (9) for 3N–H on a (0001) surface is much lower than that for 2NH$_2$+5N–H on a (1101) surface.

4. Conclusions

We devised an improved thermodynamic approach for semipolar GaN and AlN surfaces under MOVPE growth conditions. According to the improved thermodynamic analysis for semipolar surfaces, we can discuss temperature and orientation dependence of $P_{\text{Ga}}$ as well as $P_{\text{N}}$. The calculated results for $N_2$ carrier gas revealed that growth on GaN(1122) surfaces is possible at higher temperatures than on a GaN(0001) surface, while the maximum temperature for growth on an AlN(1122) surface is similar to that for growth on an AlN(0001) surface. The relationship of maximum growth temperature between (1101) and (0001) surfaces depends on the group-III element. For GaN, the maximum temperature for growth on a GaN(1101) surface is comparable to that for growth on a GaN(0001) surface for both $N_2$ and $H_2$ carrier gases. In contrast, for AlN growth on an AlN(0001) surface is possible at higher temperatures than growth on an AlN(1101) surface regardless of the group-III element. In addition, for $H_2$ carrier gas, growth on (1122) surfaces is possible at higher temperatures than for growth on (0001) surfaces for both GaN and AlN. On the other hand, for growth on (1101) surfaces, a difference is found in growth conditions between GaN and AlN surfaces. The maximum temperature for growth on GaN(1101) surfaces is comparable to that for growth on GaN(0001) surfaces, whereas the maximum temperature for growth on AlN(0001) surfaces is higher than that for growth on AlN(1101) surfaces. Furthermore, the maximum temperature for growth on (0001) surfaces is always higher than that on semipolar (1101) surfaces for both GaN and AlN. These results suggest that our analysis can be used to understand which temperatures are favorable for growth on semipolar surfaces and the dependence on surface orientation during MOVPE.

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Table I. Calculated maximum difference in growth temperature between GaN and AlN surfaces for both $H_2$ and $N_2$ carrier gases.

| Material | Orientation | Maximum temperature for $H_2$ carrier gas (°C) | Maximum temperature for $N_2$ carrier gas (°C) |
|----------|-------------|-----------------------------------------------|-----------------------------------------------|
| GaN      | (0001)      | 1025                                          | 1075                                          |
|          | (0001)      | Higher than 1300                              | Higher than 1300                              |
|          | (1122)      | 1250                                          | Higher than 1300                              |
|          | (1101)      | 1030                                          | 1105                                          |
| AlN      | (0001)      | 1530                                          | Higher than 1800                              |
|          | (0001)      | 1770                                          | Higher than 1800                              |
|          | (1122)      | 1795                                          | Higher than 1800                              |
|          | (1101)      | 1480                                          | 1650                                          |

1) S. Nakamura et al., Japan. J. Appl. Phys. 36, L1568 (1997).
2) G. Martin, A. Botchkharev, A. Rocketti, and H. Moskoc, Appl. Phys. Lett. 68, 2541 (1996).
3) F. Bernardini, V. Fiorentini, and D. Vanderbilt, Phys. Rev. B 56, R10024 (1997).
4) H. Hirayama, J. Appl. Phys. 97, 091101 (2005).
5) A. Khan, K. Balakrishnan, and T. Katona, Nat. Photon. 2, 77 (2008).
6) H. Hirayama, S. Fujikawa, N. Noguchi, J. Norimatsu, T. Takano, K. Tsukuba, and N. Kamata, Phys. Status Solidi. A 206, 1176 (2009).
7) T. Oto, R. G. Banal, K. Kataoka, M. Funato, and Y. Kawakami, Nat. Photon. 4, 767 (2010).
8) C. Pernot et al., Appl. Phys. Express 3, 061004 (2010).
9) M. Funato, K. Matsuda, R. G. Banal, R. Ishii, and Y. Kawakami, Appl. Phys. Express 5, 082001 (2012).
10) R. G. Banal, M. Funato, and Y. Kawakami, J. Cryst. Growth 311, 2834 (2009).
11) A. Rice, R. Collazo, J. Tweedie, R. Dalmau, S. Mita, J. Xie, and Z. Sitar, J. Appl. Phys. 108, 043510 (2010).
12) R. Miyagawa, S. Yang, H. Miyake, and K. Hiramatsu, Phys. Status Solidi. C 8, 2069 (2011).
13) N. Fujimoto et al., Phys. Status Solidi. C 3, 1617 (2006).
14) N. Okada et al., J. Cryst. Growth 298, 349 (2007).
15) J. Stellmach, M. Frentrup, F. Melnke, M. Pistoysveck, T. Wernicke, and M. Kneissl, J. Cryst. Growth 355, 59 (2012).
16) T. Akasaka, Y. Kobayashi, and T. Makimoto, Appl. Phys. Lett. 90, 121919 (2007).
17) N. Okada et al., J. Cryst. Growth 300, 141 (2007).
18) Y. Seta, T. Akiyama, A.-M. Pradipto, K. Nakamura, and T. Ito, J. Cryst. Growth 510, 7 (2019).
19) T. Akiyama, Y. Seta, K. Nakamura, and T. Ito, Phys. Rev. Mater. 3, 023401 (2019).
20) A. Kusaba, Y. Kangawa, P. Kempisty, K. Shiraishi, K. Kakimoto, and A. Koukitu, Appl. Phys. Express. 9, 125601 (2016).
21) A. Kusaba, Y. Kangawa, P. Kempisty, H. Valencia, K. Shiraishi, Y. Kumagai, K. Kakimoto, and A. Koukitu, Japan. J. Appl. Phys. 56, 070304 (2017).
22) J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
23) N. Troullier and J. L. Martins, Phys. Rev. B 43, 1993 (1991).
24) D. Vanderbilt, Phys. Rev. B 41, 7892(R) (1990).
25) S. G. Louie, S. Froyen, and M. L. Cohen, Phys. Rev. B 26, 1738 (1982).
26) J. Yamauchi, M. Tsukada, S. Watanabe, and O. Sugino, Phys. Rev. B 54, 5586 (1996).
27) H. Kagoshima and K. Shiraishi, Phys. Rev. B 56, 14985 (1997).
28) M. Tsukada et al., Computer Program Package TAPP (The University of Tokyo, Tokyo, 1983–2018).
29) K. Shiraishi, J. Phys. Soc. Japan 59, 3455 (1990).
30) Y. Takemoto, T. Akiyama, K. Nakamura, and T. Ito, Japan. J. Appl. Phys. 54, 085502 (2015).
31) Y. Takemoto, T. Akiyama, K. Nakamura, and T. Ito, e-J. Surf. Sci. Nanotch. 13, 239 (2015).
32) M. D. Pashley, K. W. Haberern, W. Friday, J. M. Wooldall, and P. D. Kirchner, Phys. Rev. Lett. 60, 2176 (1998).
33) Y. Kangawa, T. Akiyama, T. Ito, K. Shirasaki, and T. Nakayama, Materials 6, 3309 (2013).
34) M. R. Leys and H. Veenvliet, J. Cryst. Growth 55, 145 (1981).
35) J. Nishizawa and T. Kurabayashi, J. Electrochem. Soc. 130, 413 (1983).
36) K. Sekiguchi, H. Shirakawa, K. Chokawa, M. Araidai, Y. Kangawa, K. Kakimoto, and K. Shirasaki, Japan. J. Appl. Phys. 56, 04CJ04 (2017).
37) A. Koukitu and H. Seki, J. Cryst. Growth 189–190, 13 (1998).
38) A. Koukitu, Y. Kumagai, and H. Seki, J. Cryst. Growth 221, 743 (2000).
39) T. Yayama, Y. Kangawa, K. Kakimoto, and A. Koukitu, Phys. Status Solidi. C 7, 2249 (2010).
40) S. Zhang and S.-H. Wei, Phy. Rev. Lett. 92, 086102 (2004).
41) C. E. Dreyer, A. Janotti, and C. G. Van de Walle, Phys. Rev. B 89, 081305 (R) (2014).