

Strain-stress relationships for coherent in-plane strain in heterostructures with monoclinic crystal systems: β -($\text{Al}_x\text{Ga}_{1-x}$) $_2\text{O}_3$ on ($h0l$) β - Ga_2O_3 as example

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In this work, we derive the relationship between the states of strain and stress in pseudomorphic epitaxial layers on a substrate with differing equilibrium lattice parameters under symmetry-conserving conditions for the case of monoclinic crystal symmetry. We compare surface vectors across the template-epitaxial layer interface and impose conditions of a stress-free epitaxial layer. As a result, we demonstrate the theoretical existence of up to three possible unit cells that can establish on a given template. We demonstrate this approach for the special case of symmetry-conserving strain in a class of templates with ($h0l$) surface orientations. We use β -($\text{Al}_x\text{Ga}_{1-x}$) $_2\text{O}_3$ on ($h0l$) β - Ga_2O_3 as an example. We discuss the effects of the composition x and surface orientation on the formation of three elastically stable unit cells, their strain and stress tensors, unit-cell axes, unit-cell volumes, lattice spacings, elastic potential energies, and stress-free directions. The previous paradigm for epitaxial layer growth, where the only stress-free direction is perpendicular to the growing surface, is not generally valid for low-symmetry materials. In the example here, we find that, in addition to the stress-free growth direction for crystallographic planes with low Miller indices, two possible competing domains can exist with stress-free directions oblique to the surface of the template for almost all planes ($h0l$). We further demonstrate the effect of the orientation-dependent differently strained domains and calculate the strain-shifted anisotropic band-to-band transitions for β -($\text{Al}_{0.1}\text{Ga}_{0.9}$) $_2\text{O}_3$ on ($h0l$) β - Ga_2O_3 . We also provide examples of x-ray diffraction analyses, which may help guide the search for the occurrence of such differently strained unit cells in epitaxial layers grown by experiment, here taking the ($\bar{2}01$) surface orientation as case for demonstration.

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I. INTRODUCTION

In heteroepitaxial growth of pseudomorphic single crystalline layers on single-crystal templates, the mismatch between the respective lattice parameters of isomorphic

unit cells leads to biaxial strain in the epitaxial layers. This strain is associated with a characteristic stress distribution across the epitaxial layers. The effects of strain and stress are crucially important for understanding, for example, the modifications of thermal, electronic, optical, and optoelectronic properties. The full evaluation of the strain-stress relationships and the consequences for stable growth of single-domain epitaxial layers have not yet been elucidated for low-symmetry crystals. Low-symmetry materials, such as monoclinic β - Ga_2O_3 , and their heteroepitaxy with isovalent alloys, including bixbyite-structure In_2O_3 and corundum-structure Al_2O_3 , are of significant contemporary interest due to various emergent physical properties, such as a very large tunable band-gap energy

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[1,2]. For example, isovalent alloying of In and Al in β -(Al,Ga,In) $_2$ O $_3$ offers tuning of the band-to-band transition energies using composition- and lattice-mismatch-induced strain [3]. Monoclinic θ -Al $_2$ O $_3$ has a calculated lowest band-to-band transition of 7.2–7.5 eV [4,5], and monoclinic β -In $_2$ O $_3$ of 2.7 eV [1,6], with lattice mismatches to β -Ga $_2$ O $_3$ of 4% [7,8] and 10% [2], respectively.

Strain and stress are inherent to heteroepitaxial layer growth due to thermal expansion and lattice mismatch between heterostructure constituents. Epitaxial layer growth of β -Ga $_2$ O $_3$ has been reported on various foreign substrates [9–12] using different techniques, including metal-organic chemical vapor deposition (MOCVD) [13,14], halide vapor-phase epitaxy (HVPE) [15,16], molecular-beam epitaxy (MBE) [17,18], mist chemical vapor deposition (Mist-CVD) [19], and pulsed laser deposition (PLD) [20]. Various epitaxial relationships have been observed. For example, MOCVD permits successful deposition of ($\bar{2}01$)-oriented β -Ga $_2$ O $_3$ epitaxial layers on (0001) sapphire substrates [21]. The (201) plane of β -Ga $_2$ O $_3$ shares the same in-plane oxygen-atom arrangement as the oxygen arrangement of the (0001)-oriented sapphire template, which facilitates the growth of phase-pure (201)-oriented β -Ga $_2$ O $_3$ epitaxial layers on (0001) sapphire substrates despite the considerable lattice mismatch (6.12%) [22]. Six rotational domains with ($\bar{2}01$) orientation are often revealed in x-ray diffraction (XRD) of epitaxial layers grown on (0001) sapphire. Wang *et al.* reported MOCVD growth of high-crystalline-quality epitaxial layers of β -Ga $_2$ O $_3$ on KTaO $_3$ (KTO) substrates with the epitaxial relationships (001) β -Ga $_2$ O $_3$ parallel to KTO (001) and β -Ga $_2$ O $_3$ [001] parallel to KTO $\langle 011 \rangle$, although strain was not discussed [23]. Gallium evaporation under oxygen plasma was used by Nakagomi *et al.* to study the in-plane and out-of-plane epitaxial relationships and structural properties of β -Ga $_2$ O $_3$ epitaxial layers on (001), (111), and (110) cubic (rocksalt structure) MgO substrates. A γ -Ga $_2$ O $_3$ interface layer was observed. The authors reported and discussed the occurrence of multiple-fold rotational domain structures with different crystallographic orientations of the β -Ga $_2$ O $_3$ domains depending on the template surface orientation, while strain was not mentioned [24]. Tang *et al.* reported the growth of high-crystalline-quality ($\bar{2}01$) β -Ga $_2$ O $_3$ epitaxial layers on (001) CeO $_2$ substrates using PLD. The authors observed rotational domains with small lattice mismatch, and strain was not discussed [25]. For a discussion of strain effects and a recent summary of epitaxial layer growth of β -Ga $_2$ O $_3$ by PLD, see Mock *et al.* [26]. Nepal *et al.* reported growth of ($\bar{2}01$) β -Ga $_2$ O $_3$ epitaxial layers on (0001) 4H-SiC substrates by MBE. Domain structures and strain were not discussed [27]. Yan *et al.* studied the use of NiO buffer layers to improve the heteroepitaxial growth of β -Ga $_2$ O $_3$ using Mist-CVD. The Williamson-Hall method was used to determine microstrain from XRD investigations, which

was found to anticorrelate with the average grain size [28]. Oshima *et al.* reported the growth of (201) β -Ga $_2$ O $_3$ on (0001) sapphire using HVPE. Sixfold domain-structure properties were observed, similar to those reported from other growth methods [29].

Wen *et al.* reported epitaxial growth of (010) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ on (010) β -Ga $_2$ O $_3$ using tritertiarybutylaluminum (TTBAI) with an aluminum gas precursor in a hybrid MBE (h-MBE) system [30]. The epitaxial layers were found to be pseudomorphically grown with the highest quality. Stable layers with Al concentrations ranging from 1% to 25% were obtained. The state of strain in the epitaxial layers was not discussed. Sarker *et al.* reported on the microscopic and spectroscopic properties of (100), ($\bar{2}01$), and (010) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ epitaxial layers grown pseudomorphically on β -Ga $_2$ O $_3$ [31]. The effect of strain was not discussed. Bhuiyan *et al.* reported on growth parameters and their impacts on the surface morphology of MOCVD-grown β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ epitaxial layers on (010) β -Ga $_2$ O $_3$ substrates [32]. Strain was investigated using reciprocal-space mapping (RSM). Epitaxial layers with thickness greater than 400 nm were found to be partially relaxed, while thinner layers were fully strained. Bhuiyan *et al.* reported growth of coherently strained (001) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ epitaxial layers on (001) β -Ga $_2$ O $_3$ with x up to 25% by MOCVD. The Al concentration, crystalline structure, epitaxial-layer quality, and strain were evaluated using XRD measurements [33]. The state of strain was not quantified. Alema *et al.* reported on the growth of epitaxial layers of (010) and (100) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ on (010) and (100) β -Ga $_2$ O $_3$, respectively, using MOCVD in an N $_2$ O environment [34]. Formation of pure beta phase was found for Al concentrations up to 45%, and the epitaxial layers were found to be coherently strained up to 30%. XRD was used to evaluate the phase purity and strain characteristics. The strain parameters were not quantified. Lundh *et al.* reported on the state of strain and critical thickness of MBE-grown (010) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ epitaxial layers on (010) β -Ga $_2$ O $_3$ using XRD and RSM [35]. It was found that cracked films remained fully strained; strain parameters were not reported. Mauze *et al.* reported on metal organic catalyzed epitaxial (MOCATAXY) growth of (001) β -(Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ epitaxial layers on (001) Ga $_2$ O $_3$ [36]. RSM was used to show that the films were coherently strained. All strain parameters were quantified. Multiple domain structures were not observed.

Modifications imposed by strain onto the band-structure properties of crystalline materials can be studied using group-theory methods [37]. When lattice distortions are small compared with the original lattice parameters, a range of distortions can exist where the resulting variations of band-structure properties can be approximated as a linear perturbation of the eigenstates of the strain-free lattice. Deformation parameters can then be used to calculate the effect on a given eigenstate as a result of a

given condition of the lattice's strain or stress. The set of equations describing this relationship is also known as the strain-stress relationships for eigenstates in crystals. The structure of this set of equations is conditioned by the symmetry of the participating lattices within a given heterostructure.

Zhang *et al.* performed density functional theory analysis of hydrostatic strain-induced variations in the band structure of β -Ga₂O₃ and reported strong variation in the band-to-band transitions, anisotropy of electron mobility, and effective mass parameters [38]. The strong variations observed suggest that strain engineering could be an important approach to band-gap engineering in β -Ga₂O₃. Huang *et al.* reported on the β -to- γ phase transformation in Sn-doped and Si-implanted Ga₂O₃ as a result of local strain induced by impurity atoms that favor the formation of interstitial-divacancy complexes with subsequent phase transition [39]. Seacat, Lyons, and Peelaers predicted β -(In_{0.25}Al_{0.75})₂O₃ as a composition with lattice constants similar to β -Ga₂O₃, featuring indirect and direct band gaps of 5.96 and 5.7 eV, respectively, and a conduction-band offset of 1 eV toward β -Ga₂O₃ [40]. Barmore *et al.* investigated the effects of hydrostatic pressure up to 9 GPa on the photoluminescence shift of the R lines in Cr-doped β -Ga₂O₃ and (Al_{0.1}Ga_{0.9})₂O₃ [41]. Xu *et al.* demonstrated strain-induced phase transformation and stabilization under large template mismatch between rutile-structure α -Al₂O₃ and β -Ga₂O₃ [42]. Zhang *et al.* used density functional theory computations and discussed the effects of uniaxial, biaxial, and isotropic strain on the band-gap energy and effective mass parameters in β -Ga₂O₃ [43]. Hara *et al.* performed Vicker's indentation with Raman studies on (010) β -Ga₂O₃ and deduced deformation-potential parameters for selected modes [44]. Uchida and Sugie reported stress analysis in β -Ga₂O₃ using Raman spectroscopy and obtained phonon deformation-potential parameters in very good agreement with theoretical predictions by Korlacki *et al.* [45,46]. Hasuike *et al.* performed bending-stress investigations using Raman spectroscopy, bending (001) β -Ga₂O₃ along the [001] direction, thereby also deforming the monoclinic lattice cell [47]. The resulting deformation-potential parameters assuming monoclinic symmetry differ from those predicted for symmetry-retaining stress [46]. Mu *et al.* calculated the orientation-dependent band offsets in pseudomorphically strained and relaxed β -(Al, Ga)₂O₃/ β -Ga₂O₃ interfaces [6,7,48]. Korlacki *et al.* described the linear-perturbation-theory strain and stress relationships for optical phonon modes in monoclinic crystals for strain and stress situations that maintain the monoclinic symmetry of the crystal [46]. The relationships were extended to the Γ -point topmost-valence-band and lowest-conduction-band levels for β -Ga₂O₃ and monoclinic θ -Al₂O₃ [4].

The establishment of correct relationships between the applied stress and the resulting strain is key to correct

interpretation of the state of strain in a given heteroepitaxial or otherwise strained system. Typical approaches to deriving relationships between strain components along different crystallographic directions stem from linear elasticity theory, considering that the surface of an epitaxial film on a foreign substrate is free to expand or contract. Consequently, the stress along the growth direction is zero. This approach has been extensively used to obtain strain-stress relationships for structural, dielectric, lattice-dynamical, and electronic properties of materials with high crystal symmetries such as cubic and hexagonal [49–52]. This task is not trivial for low-symmetry crystals. Recently, Grundmann provided analytic solutions for the calculation of the strain parameters in pseudomorphically grown epitaxial layers of monoclinic symmetry onto monoclinic substrates when the growth surface is (010) as well as all (*h*0*l*) planes [53]. Numerical results for the variation of the lattice parameters and the strain energy density were shown for hypothetical growth of β -(Al, Ga)₂O₃ and β -(In, Ga)₂O₃ on β -Ga₂O₃ for lattice-mismatch-induced biaxial strain. It was assumed that the stress-free direction is aligned perpendicular to the surface. We argue here that this assumption is only valid during growth for highly symmetric crystal structures and for growth planes with low crystallographic indices. Specifically, this assumption is valid only when the determinant of the stress tensor factorizes, with the direction of growth separating from the remainder of the tensor. In this present work, it is shown that the stress-free direction in pseudomorphically grown epitaxial layers with low-symmetry crystals, in general, does not align with the template surface normal. Furthermore, we demonstrate that, in general, there are three possible unit-cell solutions that can form with different strain-stress situations under the same epitaxial conditions of an epitaxial layer growing with a fixed targeted composition, for example. Each solution has its own stress-free direction and state of elastic energy, as will be detailed in this work. Our findings are fundamentally different from all previously made considerations for the strain-stress relationships in heteroepitaxial growth of high-symmetry materials. The differently strained, stable unit-cell solutions described in this work may offer additional opportunities in the heteroepitaxial growth of low-symmetry crystals. As a result of the theory derived here, one can calculate and predict XRD patterns for multiple unit-cell solutions and search for their occurrence in experimental investigations. Example XRD patterns are shown at the end of this work, in Sec. V. We finally emphasize that in the derivation and considerations here, we deliberately consider arbitrary orientations between substrate and epitaxial layer, independent of the atomic positions at the interface, i.e., the atomic effects at the interface are not included. We also ignore effects due to reconstruction, point and extended defects, and grain boundaries.

II. THEORY

A. Interface mesh vectors

A given growth surface in a monoclinic lattice is characterized by the unit-cell vectors $\{\mathbf{a}, \mathbf{b}, \mathbf{c}\}$ and indexed using Miller indices for its crystallographic plane (hkl) . An example unit cell for monoclinic gallium oxide is shown in Fig. 1. The surface-normal vector parallels the reciprocal vector $\mathbf{g}_{hkl}$:

$$\mathbf{g}_{hkl} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^*, \quad (1)$$

with the reciprocal lattice vectors expressed as follows:

$$\mathbf{v}_i \mathbf{v}_j^* = \delta_{ij}, \quad (2)$$

where $\mathbf{v}_1 = \mathbf{a}$, $\mathbf{v}_2 = \mathbf{b}$, $\mathbf{v}_3 = \mathbf{c}$, $\mathbf{v}_1^* = \mathbf{a}^*$, $\mathbf{v}_2^* = \mathbf{b}^*$, $\mathbf{v}_3^* = \mathbf{c}^*$, and δ_{ij} is the Kronecker symbol [55].

B. Hooke's law

The linear theory of elasticity is applied here, and small-deformation stress analysis can be performed using the stress tensor σ . The stress tensor fully renders the amount of stress experienced by a unit cell within the epitaxial layer when under strain. Here, σ relates a dimensionless unit vector $\hat{\mathbf{e}}$ to its traction vector $\mathbf{T}(\hat{\mathbf{e}})$, which is a measure of the stress in units of kbar as a function of unit direction $\hat{\mathbf{e}}$:

$$\mathbf{T}(\hat{\mathbf{e}}) = \sigma \hat{\mathbf{e}}, \quad (3)$$

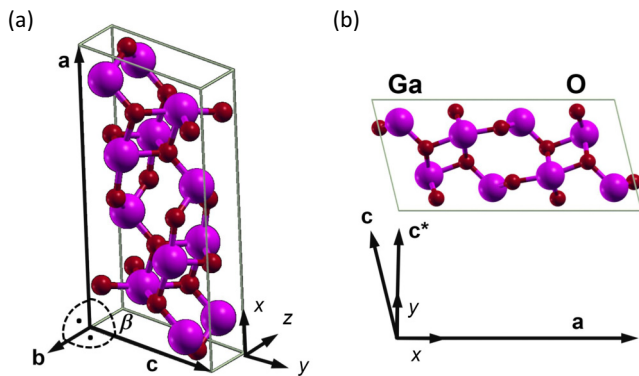


FIG. 1. (a) Unit cell of β -Ga₂O₃. Indicated are the monoclinic angle β and the Cartesian coordinate system (x, y, z) fixed to the unit cell in this work with axes $\mathbf{a}$ and $\mathbf{b}$ tied to the x and $-z$ directions, respectively. (b) View onto the $\mathbf{a}$ - $\mathbf{c}$ plane along axis $\mathbf{b}$, which points into the plane. Indicated is the reciprocal lattice vector $\mathbf{c}^*$, which is not drawn to scale. Reprinted from Ref. [54] with permission from the American Physical Society.

where σ is a symmetric, second-rank tensor in units of kbar, which has the following representation for monoclinic symmetry:

$$\sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} & 0 \\ \sigma_{xy} & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}. \quad (4)$$

The state of strain in the epitaxial layer in linear elasticity theory is fully described by the observation of all lattice vectors of the epitaxial layer without strain and with strain, $\mathbf{w}$ and $\mathbf{w}'$, respectively, and which suffice to fully define the strain tensor ϵ :

$$\mathbf{w}' - \mathbf{w} = \epsilon \mathbf{w}, \quad (5)$$

where ϵ is a symmetric, dimensionless second-rank tensor:

$$\epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ \epsilon_{xy} & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix}. \quad (6)$$

Note that the choice of the underlying coordinate systems in which the vectors $\mathbf{w}$ and $\mathbf{w}'$ are represented defines the distribution and meaning of the elements of ϵ . In this work, we chose a common coordinate system for both vectors, as shown in Fig. 2(a). Further, and without loss of generality, we choose to describe the effect of alloying—i.e., the unit-cell vector variations with composition x —such that we align the vectors $\mathbf{a}(x > 0)$ and $\mathbf{a}(x = 0)$, as shown in Fig. 2(b), and in the same coordinate system as needed to describe the vectors $\mathbf{w}$ and $\mathbf{w}'$ in Fig. 2(a).

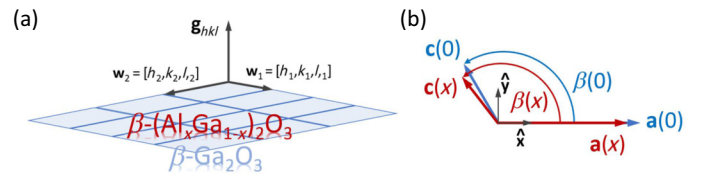


FIG. 2. (a) Definition of the mesh vectors $\mathbf{w}_{1,2}$ parallel to the interface between the β -Ga₂O₃ substrate and the $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ epitaxial layer. The reciprocal plane vector $\mathbf{g}_{hkl}$ is perpendicular to the plane (hkl) and pivotal in selecting $\mathbf{w}_{1,2}$. (b) View onto the monoclinic plane for the strain-free unit cells of the substrate ($x = 0$) and the epitaxial layer ($x > 0$) with unit-cell axes $\mathbf{a}(0)$, $\mathbf{c}(0)$, and $\mathbf{a}(x)$, $\mathbf{c}(x)$, respectively. A Cartesian coordinate system is selected, within which the changes of the unit cell due to alloying are expressed, as shown. Note that the z direction (out of the plane) is in the opposite direction to $\mathbf{b}$ (not shown here; see also Fig. 1). The strain situation discussed in this work is when the interface common to both substrate and epitaxial layer is indexed by Miller indices h, l : $(h0l)$. For the strain situation discussed in this work, the unit cell retains its monoclinic symmetry.

The generalized Hooke's law connects the strain-tensor elements with the stress-tensor elements:

$$\tilde{\sigma} = C\tilde{\epsilon}, \quad (7)$$

and we define in our previously used notation [4]

$$\tilde{\sigma} = (\sigma_{xx}, \sigma_{zz}, \sigma_{yy}, 0, \sigma_{xy}, 0)^T, \quad (8)$$

$$\tilde{\epsilon} = (\epsilon_{xx}, \epsilon_{zz}, \epsilon_{yy}, 0, 2\epsilon_{xy}, 0)^T, \quad (9)$$

where T is the transpose operator, and the tensor of the elastic constants C is given in units of kbar.

C. Epitaxial zero-stress conditions

During deposition of a crystalline layer on a crystalline substrate with a different lattice spacing than the epitaxial layer, the epitaxial layer experiences stress. It is assumed here that the thickness of the epitaxial layer is negligible compared to that of the substrate. Hence, the substrate remains stress free, and the state of stress of the epitaxial layer is homogeneous and can be fully described by σ . The scenario discussed here involves pseudomorphic growth, in which the surface of the epitaxial layer can freely expand into the half-space above the substrate without imposing any stress. Therefore, there must be one or more directions along which the epitaxial layer is free of stress.

The state of stress will depend on the amount of strain imposed onto the epitaxial layer by the mesh conditions across the interface, as well as by the internal elastic forces within the epitaxial layer, which cause further distortion of its unit cell until equilibrium is reached. When the elements of C —the elastic constants—are known, and when the strain-free unit cells of the substrate and the epitaxial layer are expressed in suitable coordinates and by known functional relationships, the strain and stress-tensor elements for a given substrate surface (hkl) and a given composition x can be calculated. Because the growing surface of the epitaxial layer is free to expand into the half-space above the surface of the substrate or to contract, the statement can be made that one or several unit direction(s) must exist, $\hat{\mathbf{v}}_0$, along which the epitaxial layer is stress free (we refer to these situations as epitaxial zero-stress conditions), hence

$$0 = \sigma \hat{\mathbf{v}}_0. \quad (10)$$

To begin with, these directions are not known. The necessary condition for any such direction to exist is that the determinant must vanish:

$$0 = \det \sigma. \quad (11)$$

A common paradigm states that the stress-free direction of an epitaxial layer is only in the direction perpendicular to

the surface; however, as shown in this work, multiple solutions of strained unit cells can exist with stress-free directions not perpendicular to the surface in low-symmetry situations, for example, in the monoclinic crystal system of β -Ga₂O₃/ β -(Al_xGa_{1-x})₂O₃.

D. Vegard's rule

The substitution of gallium by aluminum in β -(Al_xGa_{1-x})₂O₃ causes contraction of the length of all unit vectors and a small increase of the monoclinic angle [56]. This is shown schematically in Fig. 2(b). A lattice vector $\mathbf{w}(x)$ may thus be written as

$$\mathbf{w}(x) = \mathbf{w}(x=0) + \delta\mathbf{w}(x), \quad (12)$$

introducing the vector $\delta\mathbf{w}(x)$. The explicit dependence of this vector as a function of composition must be known, or described, to establish the amount and type of strain a given epitaxial layer will experience toward a given template. For example, Vegard's rule may be used to express the behaviors of the magnitudes of the unit-cell vectors, a , b , and c , and angular orientation β , as described by Kranert *et al.* [56]:

$$a(x) = a(x=0) + x\delta a, \quad (13)$$

$$b(x) = b(x=0) + x\delta b, \quad (14)$$

$$c(x) = c(x=0) + x\delta c, \quad (15)$$

$$\beta(x) = \beta(x=0) + x\delta\beta. \quad (16)$$

Here, δa , δb , δc , and $\delta\beta$ are parameters that render the linear change of the unit-cell parameters with composition x .

E. Clamping conditions

Figure 2(a) schematically depicts the mesh across a given interface characterized by crystallographic plane (hkl). The mesh may be defined by two nonparallel vectors $\mathbf{w}_{1,2}$ parallel to the interface and perpendicular to the reciprocal lattice vector $\mathbf{g}_{hkl}$:

$$\mathbf{w}_j = h_j \mathbf{a} + k_j \mathbf{b} + l_j \mathbf{c}, \quad j = 1, 2. \quad (17)$$

Miller indices $[h_j, k_j, l_j]$ may be selected such that both vectors are within the surface and perpendicular to $\mathbf{g}_{hkl}$. In this respect, $\mathbf{g}_{hkl}$ is pivotal in selecting $\mathbf{w}_j$; however, the available choices for the latter are infinite, and the most convenient may be selected.

Vectors $\mathbf{w}_{1,2}$ in Fig. 2(a) remain unchanged both in length and direction across the interface in the case of

pseudomorphic growth. Hence,

$$\mathbf{w}_j(x=0) = \mathbf{w}'_j(x>0), \quad j = 1, 2. \quad (18)$$

The latter statements can be rewritten using Eqs. (5) and (12):

$$\mathbf{w}_j(0) = [\mathbf{w}_j(0) + \delta\mathbf{w}_j(x)] + \epsilon\mathbf{w}_j(x), \quad j = 1, 2, \quad (19)$$

which highlights the requirement that the change in the lattice vector $\mathbf{w}$ due to alloying must be compensated by the amount of its strain-induced shift. Hence, two general clamping conditions can be formulated here:

$$0 = \delta\mathbf{w}_j(x) + \epsilon\mathbf{w}_j(x), \quad j = 1, 2. \quad (20)$$

These conditions state that the changes in the strain-free unit-cell vectors due to alloying must equal their corresponding vectors' negative change under the pseudomorphic strain. Note that Eqs. (10), (11), and (20) are stated within the Cartesian coordinate system of the strain-free unit cell for the epitaxial layer. The elements of ϵ bring the lattice vectors $\mathbf{w}'_j(x)$ to coincide with $\mathbf{w}_j(0)$, in length and direction [cf. Eq. (18)]. The axes of deformation expressed in the tensor elements may not coincide with any of the in-plane or out-of-plane directions of a given surface; instead, the elements of ϵ will adjust to whichever way a given unit cell is deformed under the clamping condition [57].

F. Strain-stress relationship for (*h*0*l*) interface clamping

In the present work, the approach described above is demonstrated for heteroepitaxial systems with monoclinic crystal symmetry. The cases are reduced to the situations where axis $\mathbf{b}$ is parallel to the surface; i.e., the class of surfaces characterized by (*h*0*l*) is discussed here. A generalization of this approach to triclinic cases, which includes the situation in which monoclinic epitaxial layers are distorted to triclinic symmetry, is outside the scope of the current work.

The surface vectors $\mathbf{w}_1 = [h_1, k_1, l_1]$ and $\mathbf{w}_2 = [h_2, k_2, l_2]$ are selected to be perpendicular to each other and to the surface normal:

$$\mathbf{w}_1 = l\mathbf{a} - h\mathbf{c}, \quad (21)$$

$$\mathbf{w}_2 = \mathbf{b}, \quad (22)$$

$$\mathbf{g}_{h0l} = h\mathbf{a}^* + l\mathbf{c}^*, \quad (23)$$

i.e., $k_1 = 0, h_2 = 0, l_2 = 0$. The selection $h_1 = l$ and $l_1 = -h$ places $\mathbf{w}_1$ parallel to the surface, and the scalar products vanish: $\mathbf{w}_1\mathbf{w}_2 = 0$, $\mathbf{w}_1\mathbf{g}_{h0l} = 0$, and $\mathbf{w}_2\mathbf{g}_{h0l} = 0$.

The strain-free lattice variation vectors $\delta\mathbf{w}_j$ are obtained from the difference between the strain-free vectors within the alloy for $x > 0$ and $x = 0$ (see Fig. 1):

$$\mathbf{a}(0) = a(0)\hat{x}, \quad (24)$$

$$\mathbf{b}(0) = -b(0)\hat{z}, \quad (25)$$

$$\mathbf{c}(0) = -c(0)\cos[\pi - \beta(0)]\hat{x} + c(0)\sin[\pi - \beta(0)]\hat{y}, \quad (26)$$

$$\mathbf{a}(x) = a(x)\hat{x}, \quad (27)$$

$$\mathbf{b}(x) = -b(x)\hat{z}, \quad (28)$$

$$\mathbf{c}(x) = -c(x)\cos[\pi - \beta(x)]\hat{x} + c(x)\sin[\pi - \beta(x)]\hat{y}, \quad (29)$$

$$\delta\mathbf{a}(x) = \mathbf{a}(x) - \mathbf{a}(0), \quad (30)$$

$$\delta\mathbf{b}(x) = \mathbf{b}(x) - \mathbf{b}(0), \quad (31)$$

$$\delta\mathbf{c}(x) = \mathbf{c}(x) - \mathbf{c}(0), \quad (32)$$

where $\hat{x}$ and $\hat{y}$ are unit vectors along Cartesian directions x and y within the monoclinic plane of the strain-free epitaxial layer [Fig. 2(b)].

The clamping conditions are obtained after inserting Eqs. (21) and (22) into Eq. (20) as follows:

$$0 = \epsilon[l\mathbf{a}(x) - h\mathbf{c}(x)] + [l\delta\mathbf{a}(x) - h\delta\mathbf{c}(x)], \quad (33)$$

$$0 = \epsilon\mathbf{b}(x) + \delta\mathbf{b}(x). \quad (34)$$

The task is to find all elements of ϵ . Equation (34) permits the determination of ϵ_{zz} . Because the axis $\mathbf{b}$ is parallel to the coordinate direction z and fixed within the sample surface, the only strain-tensor element related to z is ϵ_{zz} . Hence,

$$\epsilon_{zz} = \frac{b(0) - b(x)}{b(x)}. \quad (35)$$

To determine ϵ_{xx} , ϵ_{xy} , and ϵ_{yy} , Eqs. (33) and (11) must be combined. To begin with, the first clamping statement can be written as two equations:

$$0 = c_x(0)h - c_x(x)h - c_y(x)\epsilon_{xy}h - a_x(0)l + a_x(x)l + \epsilon_{xx}[-c_x(x)h + a_x(x)l], \quad (36)$$

$$0 = c_y(0)h - c_y(x)h - c_y(x)\epsilon_{yy}h + \epsilon_{xy}[-c_x(x)h + a_x(x)l]. \quad (37)$$

The latter set of equations is underdetermined and provides only two of the three strain-tensor elements. Assuming, for example, ϵ_{yy} is known, one can find closed expressions for ϵ_{xx} and ϵ_{xy} :

$$\epsilon_{xx} = \frac{-c_x(x)^2 h^2 - c_y(0)c_y(x)h^2 + c_y(x)^2 h^2 + c_y(x)^2 \epsilon_{yy} h^2}{[c_x(x)h - a_x(x)l]^2} - \frac{[a_x(0) - 2a_x(x)]c_x(x)hl + a_x(0)a_x(x)l^2 - a_x(x)^2 l^2 + c_x(0)h[c_x(x)h - a_x(x)l]}{[c_x(x)h - a_x(x)l]^2} \quad (38)$$

$$\epsilon_{xy} = h \frac{c_y(x) - c_y(0) + c_y(x)\epsilon_{yy}}{a_x(x)l - c_x(x)h}. \quad (39)$$

The elements of σ can be expressed via the elastic-tensor elements and ϵ_{xx} , ϵ_{xy} , and ϵ_{yy} ($ij = xx, xy, yy$):

$$\sigma_{ij} = C_{ij,xx}\epsilon_{xx} + C_{ij,yy}\epsilon_{yy} + C_{ij,zz}\epsilon_{zz} + 2C_{ij,xy}\epsilon_{xy}. \quad (40)$$

We can then use Eq. (11), which requires the determinant of σ to vanish. For the case of the class of $(h0l)$ surfaces of the monoclinic unit cell considered here, the condition is

$$0 = \sigma_{xx}\sigma_{yy}\sigma_{zz} - \sigma_{xy}^2\sigma_{zz}. \quad (41)$$

Inserting the expressions for ϵ_{xx} and ϵ_{xy} above into Eq. (41) leaves one equation of third order in the remaining coefficient ϵ_{yy} . Therefore, in general, the “ $\det \sigma = 0$ ” condition leads to three possible solutions. Hence, three unit cells exist that are stable from both geometrical and elastic property perspectives and that satisfy the requirement to fit the mesh defined by the vectors $\mathbf{w}_{1,2}$. Each solution renders a different strain tensor and, subsequently, differs in unit lattice vectors, stress-tensor elements, stress-free direction, unit-cell volume, total elastic energy volume density, and lattice spacing d_{h0l} along the growth direction $\mathbf{g}_{h0l}$. In the following, these different solutions will be discussed and shown for the example of a hypothetical epitaxial layer

with 10% aluminum content grown pseudomorphically on β -Ga₂O₃ with $(h0l)$ -oriented surfaces [58]. Note that our solutions are analytic and continuous with any evolution of the strain values caused by the interface mesh lattice mismatch.

G. Definition of the rotation system to address all $(h0l)$

To address a continuous variation of the template surface plane, the parameter θ is introduced such that

$$(h0l) \rightarrow (\cos \theta 0 \sin \theta) / \sqrt{2}. \quad (42)$$

Note that a factor common to both h and l can be divided out from Eqs. (36) and (37); hence, $\theta = 0, \pi/2, \pi$, etc. is equivalent to (100) , (001) , $(\bar{1}00)$, etc., respectively.

III. PARAMETER DETAILS

A. Elastic coefficients

To begin with, we provide the numerical values for the elastic coefficients for the strain-free alloy in Eq. (43) in units of kbar per unit strain, which are obtained from Ref. [4]:

$$C_{(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3} = \begin{bmatrix} 2143 + 505x & 1103 + 10x & 1200 + 90x & 0 & -197 - 76x & 0 \\ 1103 + 10x & 3300 + 546x & 669 - 80x & 0 & 119 + 24x & 0 \\ 1200 + 90x & 669 - 80x & 3248 + 748x & 0 & 75 + 71x & 0 \\ 0 & 0 & 0 & 500 + 197x & 0 & 182 + 9x \\ -197 - 76x & 119 + 24x & 75 + 71x & 0 & 689 + 329x & 0 \\ 0 & 0 & 0 & 182 + 9x & 0 & 949 + 271x \end{bmatrix}. \quad (43)$$

B. Lattice parameters

For calculation of the strain-free lattice parameters, we use the numerical results provided by Kranert *et al.* in

Ref. [56], with $a(x=0) = 12.31 \text{ \AA}$, $\delta a = -0.42 \text{ \AA}$, $b(x=0) = 3.06 \text{ \AA}$, $\delta b = -0.13 \text{ \AA}$, $c(x=0) = 5.83 \text{ \AA}$, $\delta c = -0.17 \text{ \AA}$, $\beta(x=0) = 103.72^\circ$, and $\delta \beta = 0.31^\circ$. Then,

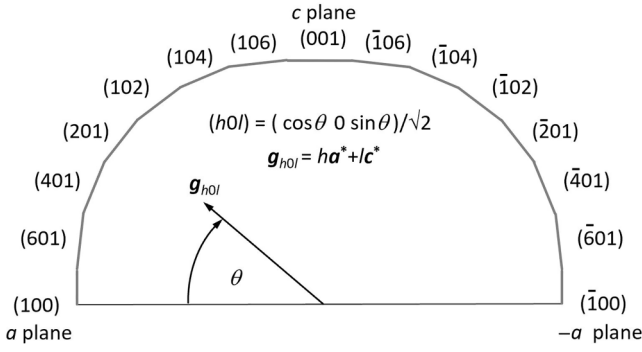


FIG. 3. Definition of rotation θ used here to parameterize the Miller indices in the surface plane ($h0l$).

Eqs. (38)–(41) can be solved either analytically or numerically.

IV. RESULTS AND DISCUSSION

A. Calculation of the strain-tensor solutions

Figure 4 depicts the four strain elements ϵ_{xx} , ϵ_{xy} , ϵ_{yy} , and ϵ_{zz} for the three unit-cell solutions for a fully strained β -(Al_{0.1}Ga_{0.9})₂O₃ epitaxial layer clamped onto a β -Ga₂O₃ template with an ($h0l$)-oriented surface. The abscissas are the rotation θ indicated in Fig. 3. Groups of three plots are shown for three differently strained unit cells found from the solution described above. The three solutions are color coded (cell #1: blue; cell #2: olive; cell #3: red), and the same color code is used in all subsequent figures unless otherwise noted. The element ϵ_{zz} is constant versus rotation and equal for all three cells. This is because the **b** axis of the epitaxial layer is clamped onto the **b** axis of the substrate and does not change, regardless of the Miller indices h and l . In Fig. 4(c), the strain ϵ_{xy} vanishes for the c -plane template because there, the **a** axis of the epitaxial layer is clamped onto the **a** axis of the substrate. Thus, there is no shear-strain element in ϵ with our coordinate-system definition. Likewise, all solutions for ϵ_{xx} pass through the same value in Fig. 4(a), which is $0.18a/a = 0.042 \text{ \AA}/12.31 \text{ \AA} = 3.3 \times 10^{-3}$.

B. Calculation of the stress-tensor solutions

Figure 5 depicts the four stress elements, σ_{xx} , σ_{xy} , σ_{yy} , and σ_{zz} for the same set of strained unit-cell solutions shown in Fig. 4. The element σ_{zz} is zero regardless of rotation for the third unit cell. This solution depicts the physically conceivable case in which the strain in the monoclinic plane is adjusted so that the unit cell of the alloyed material expands along axis **b**, such that its length coincides with that of the $x = 0$ cell. Consequently, this unit cell matches along the substrate template direction **b** and has a stress-free direction along **b**. This is consistent with

the requirement for all unit-cell solutions that **b**($x > 0$) must equal **b**($x = 0$).

It worth noting that except for σ_{xy} , there is no common crossing point. It is of further interest to note that unit cell #1 has zero stress in σ_{xx} for (100) and ($\bar{1}00$), as well as in σ_{yy} for (001), while unit cell #2 has zero stress in σ_{yy} for (100) and ($\bar{1}00$), as well as in σ_{xx} for (001). It is obvious that both cells appear to be complementary in some way; this will become clearer below. It is also noteworthy that the shear elements have common zero-crossing points at two different high-indexed Miller planes.

C. Unit-cell parameter solutions

Figure 6 depicts the magnitudes of the unit-cell lattice vectors and monoclinic angles β for the three differently strained unit cells. In accordance with the strain elements, $|\mathbf{a}|$ is common to all unit cells for the c plane, and $|\mathbf{c}|$ is common for (100) and ($\bar{1}00$). Additionally, $|\mathbf{b}|$ is constant for all cells, regardless of surface orientation h and l . The variations of the lattice-vector lengths reflect the possible changes among the four lattice parameters to adapt to the respective template by maintaining the clamping conditions and by satisfying the stress-free condition.

D. Unit-cell volume, d_{hkl} , elastic unit-cell energy, and stress-free directions

Moving forward, a differentiation is necessary to identify which of the three cells are most, more, or not likely at all to develop during epitaxial growth. Figure 7(a) shows the volumes of the three differently strained unit cells versus rotation, together with the volume of the strain-free unit cell of β -(Al_{0.1}Ga_{0.9})₂O₃. The unit-cell volume is calculated using

$$V = \mathbf{a} \cdot (\mathbf{b} \times \mathbf{c}), \quad (44)$$

where $\times$ is the cross product. In accordance with the behavior of strain elements, which are overall largest for cell #3 and smallest for cell #1, the unit-cell volume is largest for the least strained cell #1 and smallest for the most strongly strained cell #3. The same property is reflected by the behavior of the lattice-plane distances along the surface normal, d_{hkl} :

$$d_{hkl} = \frac{\sin \beta}{\sqrt{\frac{h^2}{a^2} + \frac{k^2 \sin^2 \beta}{b^2} + \frac{l^2}{c^2} - \frac{2hl \cos \beta}{ac}}}. \quad (45)$$

Figure 7(b) shows the differences in lattice spacing for the three differently strained lattices of the epitaxial layer versus rotation relative to d_{h0l} of the strain-free lattice of β -(Al_{0.1}Ga_{0.9})₂O₃. The lattice spacing is identical for the directions with the lowest Miller indices, (100), (001) and ($\bar{1}00$), and it is largest for planes (101) and ($\bar{1}01$). The cell

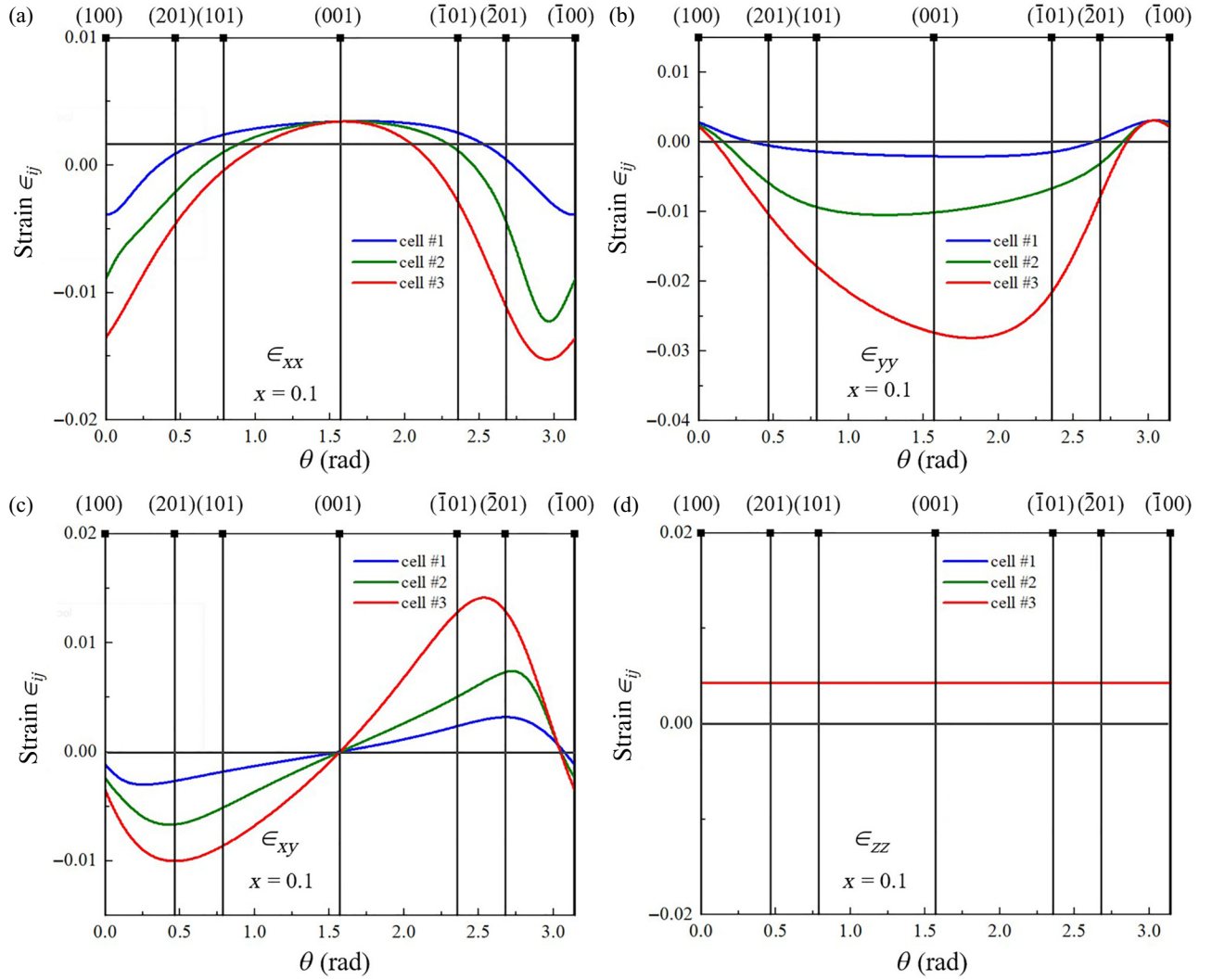


FIG. 4. Strain elements (a) ϵ_{xx} , (b) ϵ_{yy} , (c) ϵ_{xy} , and (d) ϵ_{zz} for a fully strained β -($\text{Al}_{0.1}\text{Ga}_{0.9}$) $_2\text{O}_3$ epitaxial layer clamped onto a β - Ga_2O_3 template with an $(h0l)$ -oriented surface. Rotation θ is indicated in Fig. 3. Three differently strained elastically stable unit-cell solutions are found here (cell #1: blue; cell #2: olive; cell #3: red). Note that all three unit cells have constant strain ϵ_{zz} along the $\mathbf{b}$ axis; therefore, the blue and olive lines are hidden under the flat red line in panel (d).

with largest strain and smallest volume also reflects the largest changes in overall lattice spacing. The amount of elastic potential energy stored within the strained unit cells per unit displacement can be calculated from the strain- and stress-tensor elements:

$$u = \frac{1}{2} \sum_{i=1}^6 \tilde{\epsilon}_i \tilde{\sigma}_i, \quad (46)$$

$$u = \frac{1}{2} \sum_{i=1}^6 \sum_{j=1}^3 C_{ij} \tilde{\epsilon}_i \tilde{\epsilon}_j, \quad (47)$$

and $\tilde{\sigma}_i$ and $\tilde{\epsilon}_i$ are defined in Eqs. (8) and (9), respectively. Figure 7(c) shows u for the three differently strained lattices of the epitaxial layer versus rotation. As expected,

cell #1, which has the lowest overall strain, reflects the lowest elastic potential energy, while cell #3 reveals a very large potential energy. It is thus clear that cell #1 is most likely to form during epitaxial growth, while cells #2 and #3 will require much higher energy to form; however, in addition to the elastic potential energy, it is also important to evaluate the stress-free directions for every unit-cell solution. The requirement that the determinant vanishes for the stress tensor was only the necessary condition for a stress-free cell to exist. The final question to answer is in which direction the zero stress within a given unit cell is pointing. Since the determinant of σ vanishes for all three unit cells, for each, there must be at least one direction $\mathbf{v}_{0,j}$ along which the stress is zero:

$$\sigma \mathbf{v}_{0,j} = 0, \quad j = 1, 2, 3. \quad (48)$$

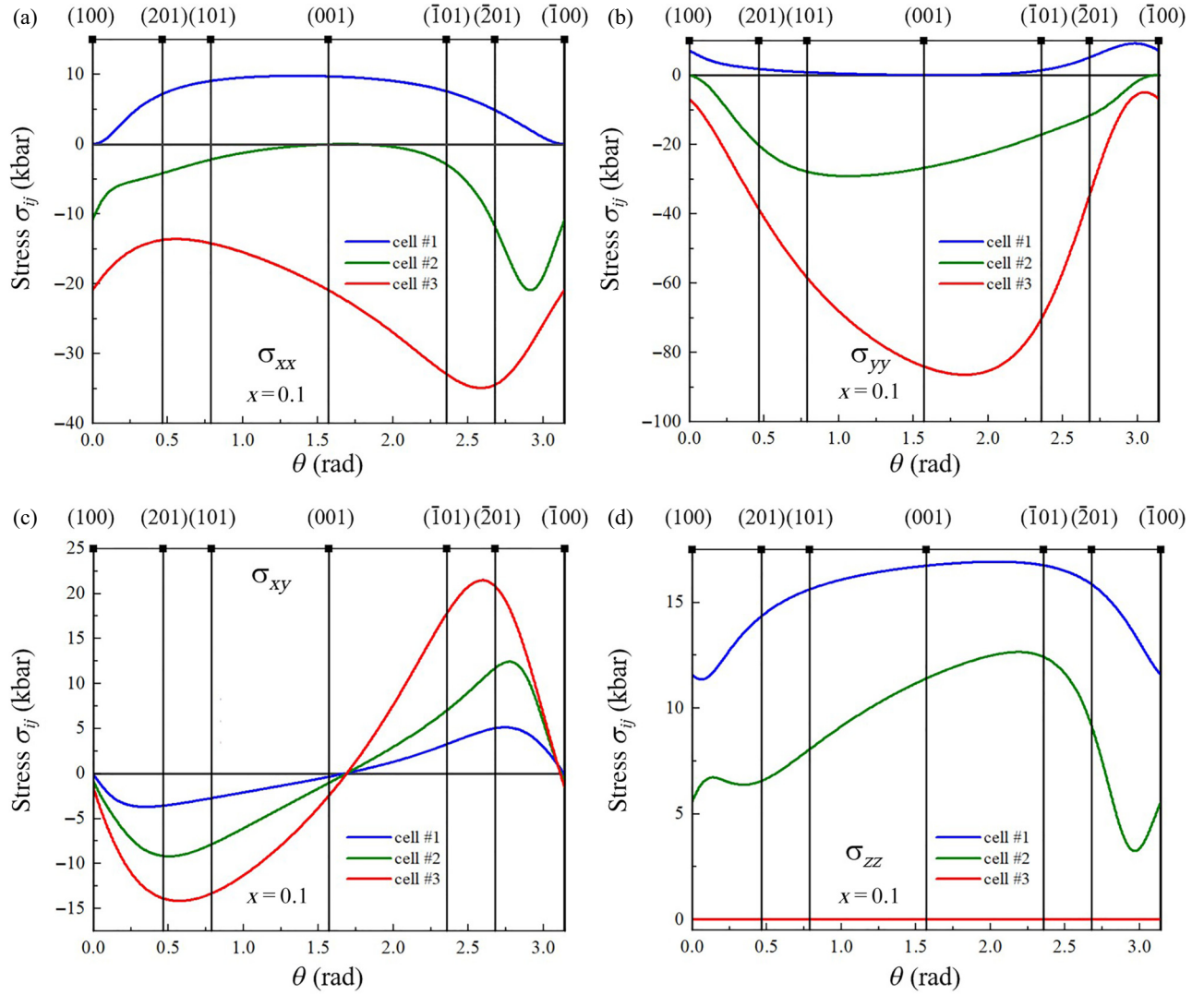


FIG. 5. Same as Fig. 4 for the stress-tensor elements. Note that while all unit-cell solutions found here always have constant strain ϵ_{zz} along axis **b**, one solution (cell #3) has zero stress along axis **b**, while the other two show varying stress along **b**. Cell #3 is strained within the monoclinic plane to the extent that its axis **b** is equal in length with the substrate, and its unit cell's stress-free direction is then parallel to the substrate. See also Fig. 7(d).

Inspection of Fig. 5(d) reveals that for unit cell #3, element σ_{zz} is zero throughout the rotation. Figure 6(d) reveals that the length of axis **b** is also constant for unit cell #3. Hence, $\mathbf{v}_{0,3}$ is parallel to direction *z*—that is, parallel to the surface—regardless of *h* and *l*. The stress-free direction being parallel to *z* in this scenario means that one can deform the unit cell of the epitaxial layer along $\mathbf{w}_1$ and along the surface normal until the internal elastic forces contract the length along axis **b** such that the latter matches the axis of the substrate. The stress-free directions for cell #1 and cell #2 can be calculated from the data shown in Fig. 5. The vectors $\mathbf{v}_{0,j}$, $j = 1, 2$ are

$$\mathbf{v}_{0,j} = -\sigma_{xx,j}\hat{x} + \sigma_{xy,j}\hat{y} = -\sigma_{xy,j}\hat{x} + \sigma_{yy,j}\hat{y}, \quad (49)$$

where $\sigma_{\dots,j}$ indicates elements of the stress tensor of the strained unit cell $j = 1, 2$. Hence, the vectors $\mathbf{v}_{0,j}$ are expressed within the unit cell of the strained epitaxial layer and within the coordinates shown in Fig. 2. The angle between the stress-free direction and the surface vector, $\mathbf{g}_{h0l}$, $\cos^{-1}(\mathbf{g}_{h0l}\mathbf{v}_{0,j}/g_{h0l}v_{0,j})$ is plotted in Fig. 7(d). Note that the vector $\mathbf{g}_{h0l}$ is different for each of the strained unit-cell solutions discussed here because the vectors **a** and **c** differ for each cell, as shown in Fig. 6; however, the epitaxial layer is lined up with the template such that $\mathbf{g}_{h0l}$ between substrate and layer are parallel to each other. This is ensured because of the clamping conditions, which require that the vectors $\mathbf{w}_{1,2}$ line up between the substrate and the epitaxial layer. Accordingly, the vectors **a** and **c** change via the respective unit-cell strain tensors for each

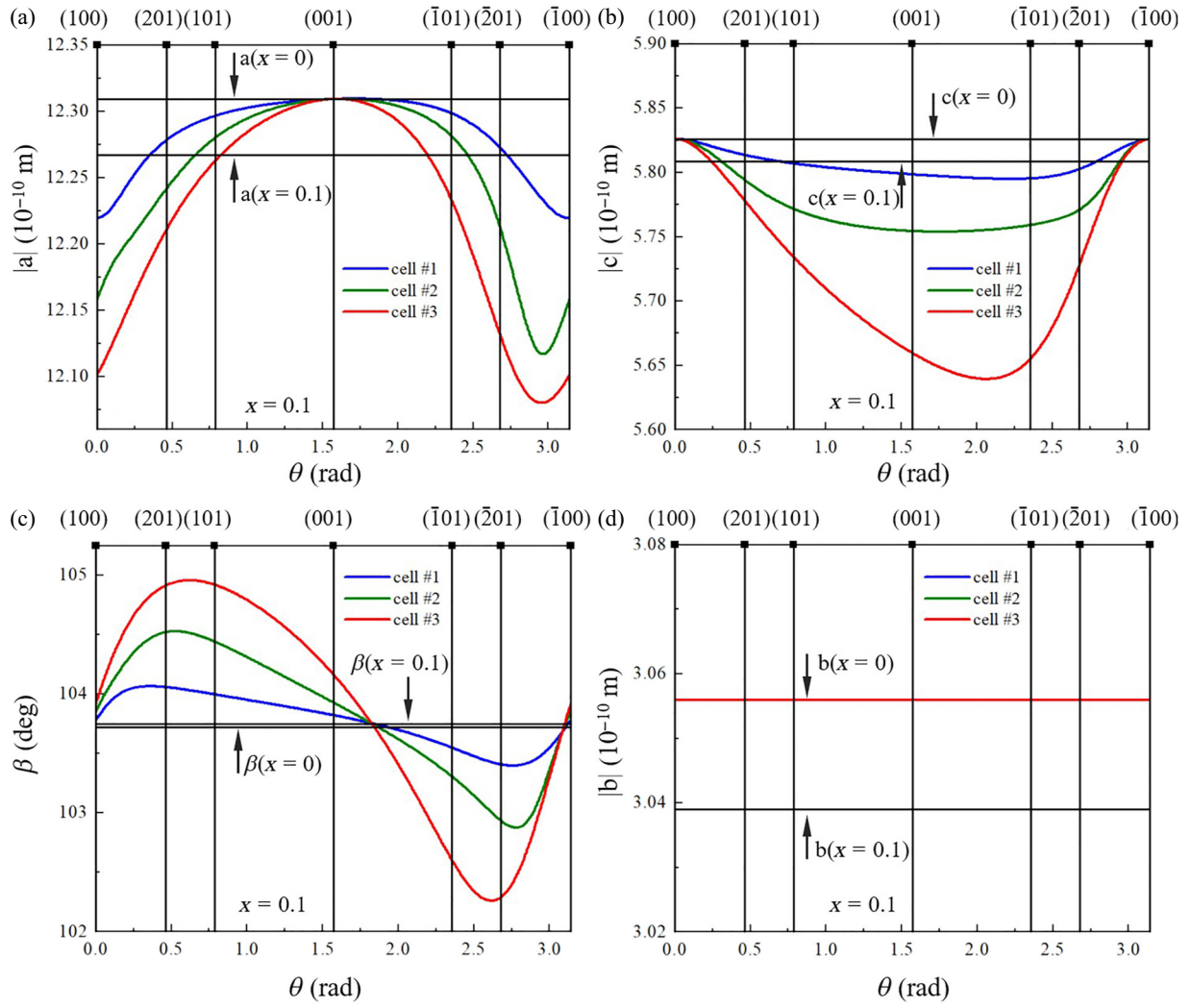


FIG. 6. Same as Fig. 4 for the magnitudes of the epitaxial layer unit-cell lattice vectors [(a) $|a|$; (b) $|c|$; (d) $|b|$] and (c) monoclinic angle β . Values for the strain-free β -Ga₂O₃ substrate ($|a| = 12.3092$ Å, $|b| = 3.05597$ Å, $|c| = 5.82579$ Å, $\beta = 103.719^\circ$) and strain-free β -(Al_{0.1}Ga_{0.9})₂O₃ ($|a| = 12.2672$ Å, $|b| = 3.03897$ Å, $|c| = 5.80879$ Å, $\beta = 103.75^\circ$) are indicated by horizontal lines. Note that values for $|b|$ are constant and equal for all unit cells, and they coincide with the strain-free value for $x = 0$. Therefore, the olive and blue lines for unit cells #1 and #2, and the horizontal line for $|b|(x = 0)$, are hidden under the flat red line for unit cell #3 in panel (d). The strain-free value for β -(Al_{0.1}Ga_{0.9})₂O₃ is smaller; hence, the strain-tensor element ϵ_{zz} is positive, constant, and equal for all unit cells. See Fig. 4(d).

unit-cell solution. Hence, $\mathbf{g}_{h0l}$ in every cell also renders the surface normal for the epitaxial layer. As can be seen in Fig. 7(d), cell #1 has its stress-free direction close to the surface normal, while that of cell #2 is mostly parallel to the surface; however, except for two specific surfaces ($h0l$), both directions deviate from the surface normal and from being parallel to the surface.

We can therefore conclude that first, the stress-free direction is not always perpendicular to the surface—in fact, it differs from the surface normal for most of the ($h0l$) planes discussed here. Second, the second cell solution also has its stress-free direction different from the surface

normal. Hence, the second cell could also grow, albeit requiring higher densification due to the reduced lattice spacing and the smaller unit-cell parameters. Therefore, unit cell #2 will require higher elastic potential deformation energy. Nonetheless, during nonequilibrium growth, such as in metal-organic vapor-phase epitaxy (MOVPE) or MBE, the formation of competing domains with different unit-cell dimensions is possible. Third, an unlikely yet possible domain may form, with a much higher elastic-energy requirement, whose stress-free direction in the class of ($h0l$) surfaces aligns parallel to the surface regardless of the choice of h and l .

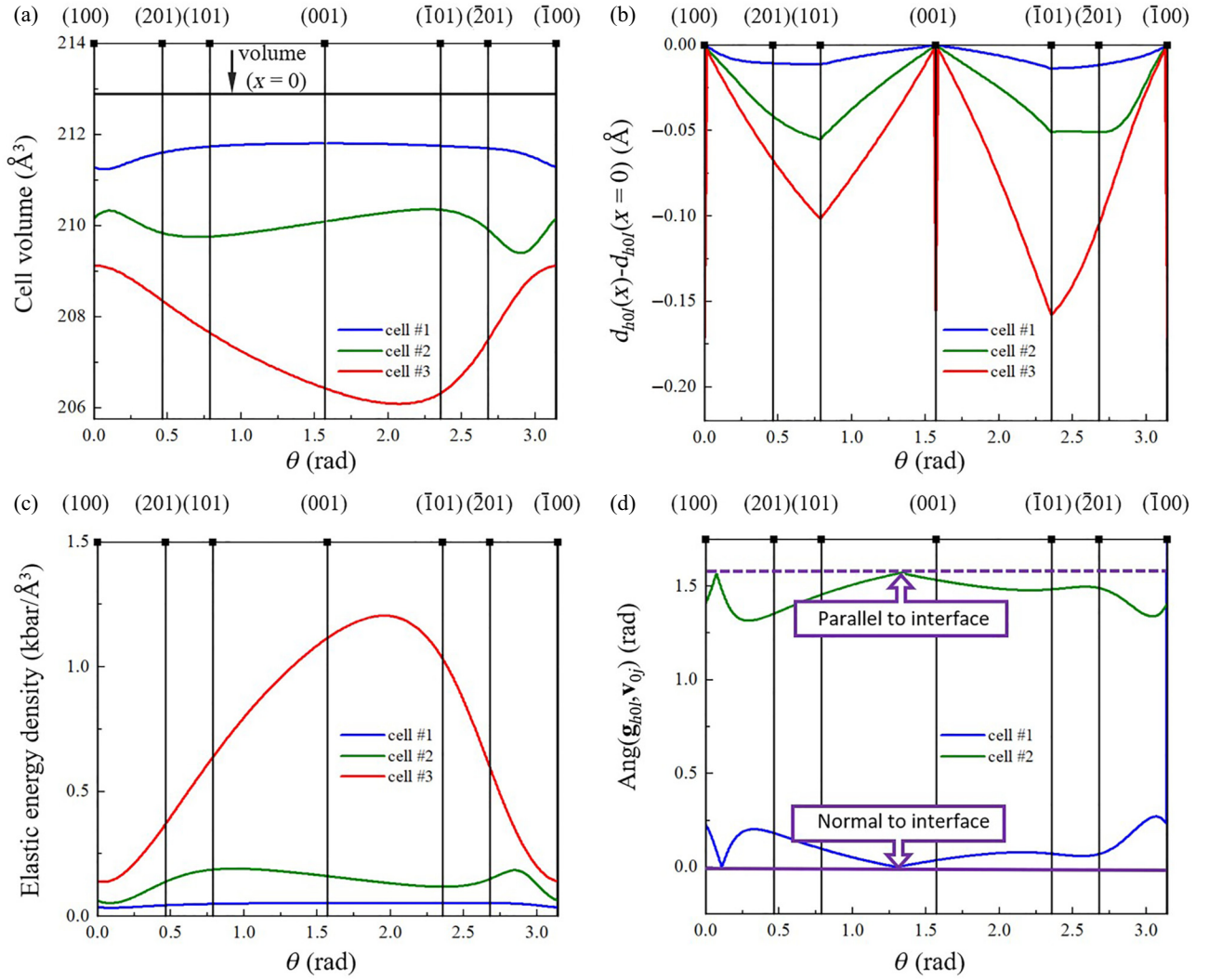


FIG. 7. Same as Fig. 4 for (a) unit-cell volume, (b) the change in lattice spacing in the growth direction, $d_{h0l}(x = 0.1) - d_{h0l}(x = 0)$, (c) elastic energy density, and (d) the stress-free directions. Note that the third unit-cell stress-free direction (red line) is parallel to axis **b** regardless of rotation θ and is not shown in panel (d). The stress-free directions are indicated by the angular difference between the stress-free direction in the Cartesian coordinate system of the unit cell in Fig. 2(b) and the rotation θ . When the difference is zero, the stress-free direction points along the surface normal, while when the difference is $\pm\pi/2$, the direction lies exactly within the surface. As can be seen, for most surfaces, the stress-free directions point obliquely to the surface. Accordingly, cell #1 and cell #2 are possible competing domains during growth.

E. Further symmetry considerations

The approach presented here is generally applicable to higher symmetries. In principle, the approach is also applicable, upon additional modifications, to triclinic symmetry cases, which shall be considered in a forthcoming work. Furthermore, in cases in which a monoclinic symmetry template has none of its lattice unit-cell axes aligned within its (hkl) surface plane, the biaxial strain introduced by the interface mesh conditions will reduce the symmetry of the epitaxial layer to triclinic. All shear strain and all shear stress elements must then be considered in ϵ and σ . All three unit cells may then compete during epitaxial growth, with stress-free directions that are no

longer strictly parallel or perpendicular to the template surface.

F. Band-to-band transitions in pseudomorphically strained $(\text{Al}_{0.1}\text{Ga}_{0.9})_2\text{O}_3/(\text{h}0\text{l})\text{Ga}_2\text{O}_3$

We use the obtained strain-stress relationship for the $(h0l)$ surfaces and calculate the effect of the resulting strain in the hypothetical pseudomorphically strained epitaxial $\beta\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$ layer on the three lowest band-to-band transitions. We employ the approach proposed by Korlacki *et al.* [4], which uses Vegard's rule. Using deformation potentials for $\beta\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$ as described by Korlacki *et al.* in Ref. [4], the following set of equations emerges

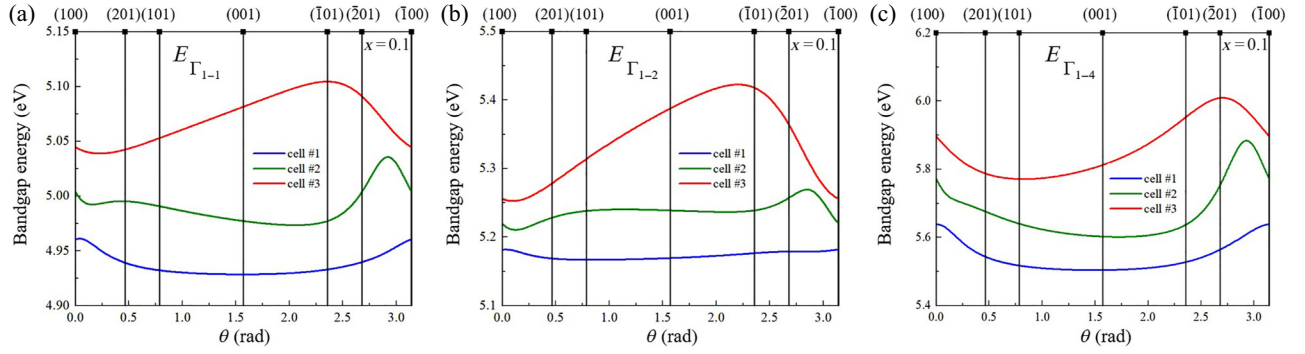


FIG. 8. Same as Fig. 4 for the band-to-band transitions in the strained hypothetical β -(Al_{0.1}Ga_{0.9})₂O₃ epitaxial layer. (a) Transition energy between Γ -point conduction band level c_1 and valence band level v_1 , (b) c_1 - v_2 , and (c) c_1 - v_4 . The deformation potential parameters used for β -(Al_xGa_{1-x})₂O₃ are taken from Korlacki *et al.* [4].

for the combined strain and composition dependencies for transitions between Γ -point conduction-band level c_1 and valence-band levels v_1 , v_2 , and v_4 ($E_{\Gamma_{1-1}}$, $E_{\Gamma_{1-2}}$, and $E_{\Gamma_{1-4}}$):

$$E_{\Gamma_{1-1}}(\text{eV}) = 5.04 + 2.613x \\ + (-8.38 - 0.90x)\epsilon_{xx} + (0.346 + 0.22x)\epsilon_{xy} \\ + (-5.93 - 1.23x)\epsilon_{yy} + (-9.56 - 1.71x)\epsilon_{zz} \\ + b_{\Gamma_{1-1}}x(x-1), \quad (50)$$

$$E_{\Gamma_{1-2}}(\text{eV}) = 5.40 + 2.594x \\ + (-7.75 - 0.63x)\epsilon_{xx} + (2.29 + 1.54x)\epsilon_{xy} \\ + (-8.49 - 1.46x)\epsilon_{yy} + (-7.74 - 1.01x)\epsilon_{zz} \\ + b_{\Gamma_{1-2}}x(x-1), \quad (51)$$

$$E_{\Gamma_{1-4}}(\text{eV}) = 5.64 + 2.648x \\ + (-8.43 + 0.76x)\epsilon_{xx} + (3.34 - 0.52x)\epsilon_{xy} \\ + (-11.9 - 3.41x)\epsilon_{yy} + (-6.33 - 0.98x)\epsilon_{zz} \\ + b_{\Gamma_{1-4}}x(x-1), \quad (52)$$

where x is the aluminum concentration and for convenience, we ignore the bowing parameters $b_{\Gamma_{1-1}} = 0$, $b_{\Gamma_{1-2}} = 0$, and $b_{\Gamma_{1-4}} = 0$. The latter are nonzero in reality, as discussed by Korlacki *et al.* in Ref. [4], but they are small and depend on ordering and higher-order lattice perturbations. Their effect may be neglected here for the purpose of demonstrating the strain effects. The results of the strain-induced variations of the transition energies for the three different cells are shown for fully strained β -(Al_xGa_{1-x})₂O₃ in Fig. 8. As can be seen, the band-to-band transitions vary strongly across the different crystallographic surface orientations, and the shifts are stronger with increasing cell index. The latter is expected because the overall strain is largest for the cell with the strongest lattice distortions. It is also noted that all transitions are blue-shifted, regardless of orientation and for all cells.

V. XRD EXAMPLES

In this section, Bragg's equation is used to demonstrate the effects of the three differently strained elastically stable unit cells for the example of a hypothetical epitaxial layer with 10% aluminum content grown pseudomorphically on β -Ga₂O₃ with ($h0l$)-oriented surfaces. For any given lattice spacing d_{hkl} in Eq. (45) and an x-ray beam with wavelength $\lambda_{\text{Cu-K}\alpha,1} = 1.5406 \text{ \AA}$,

$$\theta_{hkl} = \sin^{-1} \left(\frac{n\lambda_{\text{Cu-K}\alpha,1}}{2d_{hkl}} \right), \quad (53)$$

the diffraction (scattering) condition is obtained at angle θ_{hkl} , where n is the diffraction order (Fig. 9). An Ewald sphere can be constructed to produce maps of geometric diffraction conditions, ignoring amplitude and atomic-form-factor considerations here for simplicity. In this way, one can identify the conditions under which diffraction spots occur and compare those for the three differently strained unit cells discussed in this work. In addition, one can compare these with the diffraction spots of the same lattice planes due to the substrate as well as a hypothetical fully relaxed (strain-free) epitaxial layer of the same aluminum composition. To begin, a reference lattice plane ($h'k'l'$) may be selected to represent the polar plane of the Ewald sphere. The polar axis of this sphere is then characterized by the surface normal, which is parallel to the reciprocal vector $\mathbf{g}_{h'k'l'}$. Then, three orthogonal vectors can be selected, ($\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$), which form a right-handed Cartesian coordinate system such that

$$\mathbf{u}_3 = \mathbf{g}_{h'k'l'}, \quad (54)$$

$$\mathbf{u}_2 = \mathbf{u}_3 \frac{\mathbf{b} \cdot \mathbf{u}_3}{\mathbf{u}_3 \cdot \mathbf{u}_3} - \mathbf{b}, \quad (55)$$

$$\mathbf{u}_1 = \mathbf{u}_2 \times \mathbf{u}_3. \quad (56)$$

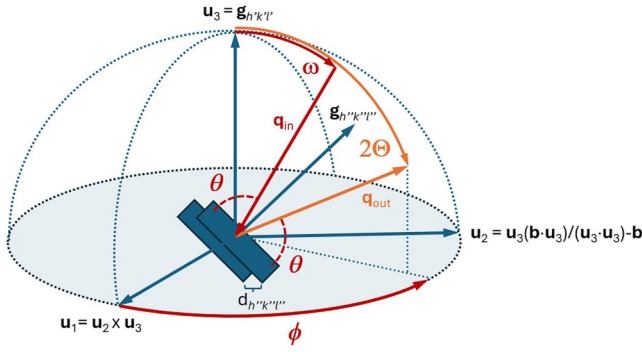


FIG. 9. Definition of the coordinate system used for calculations of the XRD lattice diffraction conditions for monoclinic β -Ga₂O₃ substrates with a coherently strained (Al_xGa_{1-x})₂O₃ epitaxial layer. The incident direction of the x-ray beam with wavelength $\lambda_{\text{Cu-K}\alpha,1}$ is indicated by $\mathbf{q}_{\text{in}}$. The scattered x-rays are collected in the direction $\mathbf{q}_{\text{out}}$. The x-ray beams share a common plane with the reciprocal lattice vectors $\mathbf{g}_{h'k'l'}$ and $\mathbf{g}_{h''k''l''}$, where $\mathbf{g}_{h'k'l'}$ is the reference vector in the Ewald-sphere presentation, and $(h''k''l'')$ indicates the plane whose lattice spacing $d_{h''k''l''}$ [Eq. (45)] satisfies the Bragg condition [Eq. (53)]. The reciprocal vector $\mathbf{g}_{h''k''l''}$ is normal to the plane $(h''k''l'')$. Incident and refracted x-rays are recorded versus incident angle ω and reflected angle 2Θ . The polar coordinate ϕ addresses the rotation of the plane of incidence around $\mathbf{g}_{h'k'l'}$. The coordinate axes $(\mathbf{u}_1, \mathbf{u}_2, \mathbf{u}_3)$ for the Ewald sphere are constructed from the choice of the reference vector $\mathbf{g}_{h'k'l'}$ and lattice axis $\mathbf{b}$. Note that for the cases discussed in this work—i.e., pseudomorphically grown epitaxial layers onto template surfaces indexed with $(h, 0, l)$ planes—the $\mathbf{b}$ axes are always identical between substrate and epitaxial layer. See Fig. 1 for unit-cell axes definitions.

Note that we select $\mathbf{u}_2$ such that this unit vector represents the direction of the projection of the β -Ga₂O₃ lattice axis $\mathbf{b}$ onto the plane $(h'k'l')$. Also note that in the cases considered in this work—i.e., epitaxial strain on templates with $(h0l)$ -oriented surfaces—the directions and magnitudes of all axes $\mathbf{b}$ in the substrate and in the epitaxial layers are the same. Then, the incident angle ω of the incident x-ray beam in direction $\mathbf{q}_{\text{in}}$ relative to the scattering-zone vector $\mathbf{g}_{h'k'l'}$ for diffraction on the plane (h'', k'', l'') is obtained by

$$\omega = \frac{\cos^{-1}(\mathbf{g}_{h'k'l'} \cdot \mathbf{g}_{h''k''l''})}{\sqrt{(\mathbf{g}_{h'k'l'} \cdot \mathbf{g}_{h'k'l'}) (\mathbf{g}_{h''k''l''} \cdot \mathbf{g}_{h''k''l''})}} + \theta_{h''k''l''} - \frac{\pi}{2}. \quad (57)$$

Then, the reflected angle 2Θ of the scattered x-ray beam in the direction $\mathbf{q}_{\text{out}}$ relative to the scattering-zone vector $\mathbf{g}_{h'k'l'}$ for diffraction on the plane (h'', k'', l'') is obtained by

$$2\Theta = \frac{\cos^{-1}(\mathbf{g}_{h'k'l'} \cdot \mathbf{g}_{h''k''l''})}{\sqrt{(\mathbf{g}_{h'k'l'} \cdot \mathbf{g}_{h'k'l'}) (\mathbf{g}_{h''k''l''} \cdot \mathbf{g}_{h''k''l''})}} - \theta_{h''k''l''} + \frac{\pi}{2}. \quad (58)$$

The polar coordinate ϕ is expressed relative to the projection of the axis $\mathbf{b}$ onto the plane (h', k', l') ; i.e., $\phi = 0$ when

$\mathbf{g}_{h''k''l''}$ is within the plane $(\mathbf{u}_1, \mathbf{u}_3)$ and $\phi = \pm\pi/2$ when $\mathbf{g}_{h''k''l''}$ is within the plane $(\mathbf{u}_2, \mathbf{u}_3)$ (see also Fig. 9). Hence,

$$\phi = \tan^{-1} \left(\frac{\mathbf{g}_{h''k''l''} \cdot \mathbf{u}_2}{\sqrt{\mathbf{u}_2 \cdot \mathbf{u}_2}} \right), \quad (59)$$

where a different choice for $\mathbf{u}_2$ is only needed when $\mathbf{g}_{h''k''l''}$ is selected to be equal to the reciprocal axis $\mathbf{g}_{h'k'l'} = \mathbf{b}^* = \mathbf{b} = \mathbf{g}_{010}$.

Scattering conditions are found for planes (h'', k'', l'') in Eq. (53). Then, after choosing $\mathbf{g}_{h''k''l''}$, corresponding values for ω and ϕ are calculated and displayed as polar coordinates $(r, \phi) \rightarrow (\omega, \phi)$ or in coordinates projected onto the plane $(\mathbf{u}_1, \mathbf{u}_2) \rightarrow (\mathbf{x}, \mathbf{y})$ with coordinates $(\sin \omega \cos \phi, \sin \omega \sin \phi)$. Computations are repeated for strain-free β -Ga₂O₃, strain-free β -(Al_{0.1}Ga_{0.9})₂O₃, and for the three strained unit cells discussed in this work. For the latter, the orientation of the β -Ga₂O₃ substrate—i.e., the orientation of the template—must be selected because the strain varies with the template's surface orientation. This was discussed at great length in the previous sections. Here, for the purpose of demonstration only, and without loss of generality, we select the situation of the (201) surface, which corresponds to $\theta \approx 2.6766$ rad in Eq. (42).

Figure 10 presents a summary of all lattice diffraction conditions for β -Ga₂O₃ (solid blue symbols) for reflections collected when the plane $(\bar{2}22)$ is selected as the polar plane. This is the plane around which the diffraction angles ω and 2Θ are adjusted relative to the surface normal of the plane (Fig. 9). The lattice indexes run from (h'', k'', l'') with $h'' = -5, \dots, 5$, $l'' = -5, \dots, 5$, and $k'' = -5, \dots, 5$. Included are the diffraction peaks under the same conditions for a hypothetical strain-free β -(Al_{0.1}Ga_{0.9})₂O₃ epitaxial layer (red solid symbols), and the three strained unit cells discussed in this work for pseudomorphic growth on $(\bar{2}01)$ β -Ga₂O₃ templates (cell #1: open blue circles, cell #2: open olive circles, cell #3: open red circles). Note that the strain-free layer has its axes $\mathbf{a}$ and $\mathbf{b}$ parallel to those of the β -Ga₂O₃ substrate [see Eqs. (27) and (28)]. Only first-order diffraction conditions ($n = 1$) are shown. It can be seen that the locations of the diffraction conditions differ, in general, between the substrate, the strain-free layer, and the differently strained epitaxial layers. This difference also varies among different lattice directions. Figure 11 depicts a small section from Fig. 10 to better visualize the variations among the diffraction conditions. This selection has been made without any further reasoning, except for the purpose of demonstration. Diffraction peaks for $(4\bar{3}0)$, $(5\bar{3}0)$, $(0\bar{3}2)$, $(3\bar{3}1)$, $(4\bar{3}1)$, and $(5\bar{3}1)$ planes are displayed in this detail section of the Ewald sphere. It can be seen that, for all diffraction conditions displayed, the three differently strained unit cells cause diffraction that fall along apparently straight lines together

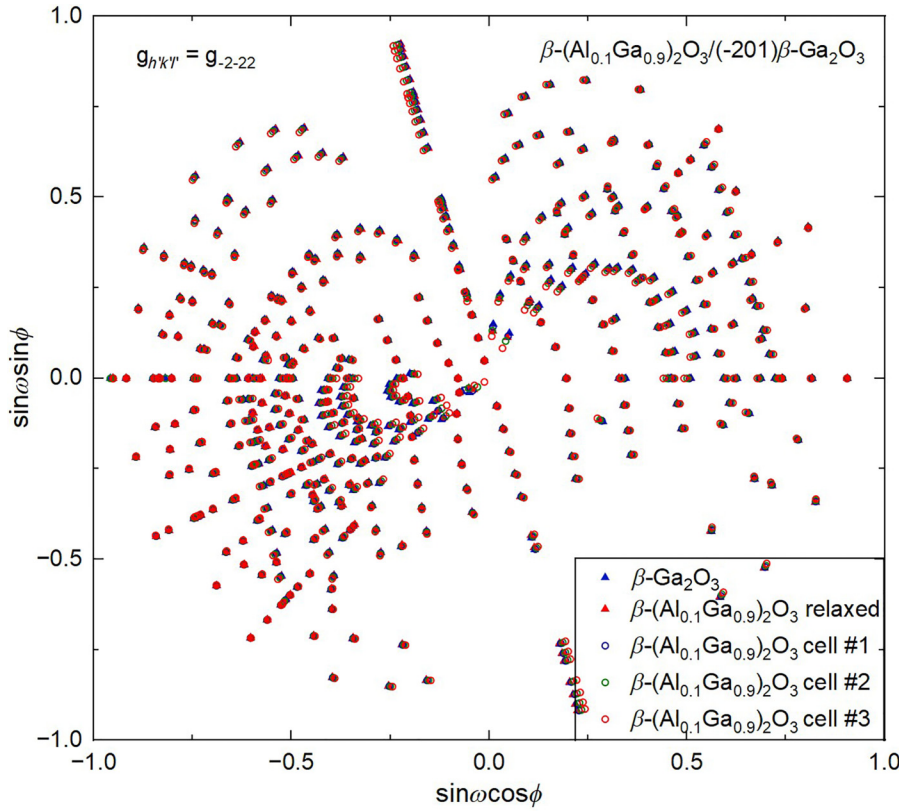


FIG. 10. Presentation of lattice diffraction conditions for β -Ga₂O₃ (solid blue symbols), strain-free β -(Al_{0.1}Ga_{0.9})₂O₃ (solid red symbols), and the three strained unit cells discussed in this work for pseudomorphic growth on $(\bar{2}01)$ β -Ga₂O₃ templates (cell #1: open blue circles, cell #2: open olive circles, cell #3: open red circles). Lattice indexes run from (h'', k'', l'') with $h'' = -5, \dots, 5$, $l'' = -5, \dots, 5$, and $k'' = -5, \dots, 5$. The incident angle (ω) and polar angle (ϕ) are defined in Fig. 9. The reference vector chosen in this example is $\mathbf{g}_{h'k'l'} = \mathbf{g}_{\bar{2}\bar{2}2}$. The strain situation is that for 10% aluminum content in the pseudomorphically grown epitaxial layer on a $(\bar{2}01)$ -oriented β -Ga₂O₃ template. The diffraction order is $n = 1$.

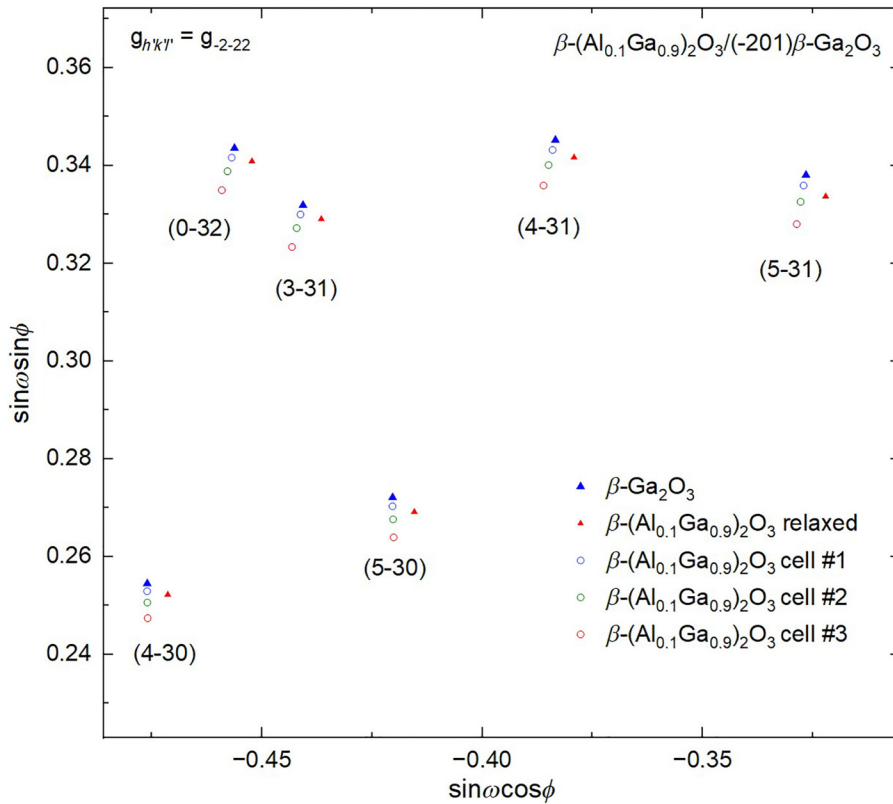


FIG. 11. Same as Fig. 10 with detail for selected diffraction peaks. Note the formation of straight lines for each reflection with respect to the diffraction peaks from the β -Ga₂O₃ template and the three coherently strained epitaxial layers, while the diffraction peaks from the hypothetically assumed strain-free, i.e., fully relaxed, (Al_xGa_{1-x})₂O₃ epitaxial layer are shifted to different locations. The straight lines are caused by the common mesh planes at the interface due to coherent growth, serving as a proof of the concept that the results obtained in this work correspond to three differently strained unit cells with different lattice constants but the same interface mesh conditions. These diffraction peaks can be measured using reciprocal-space maps or single 2theta-omega scans.

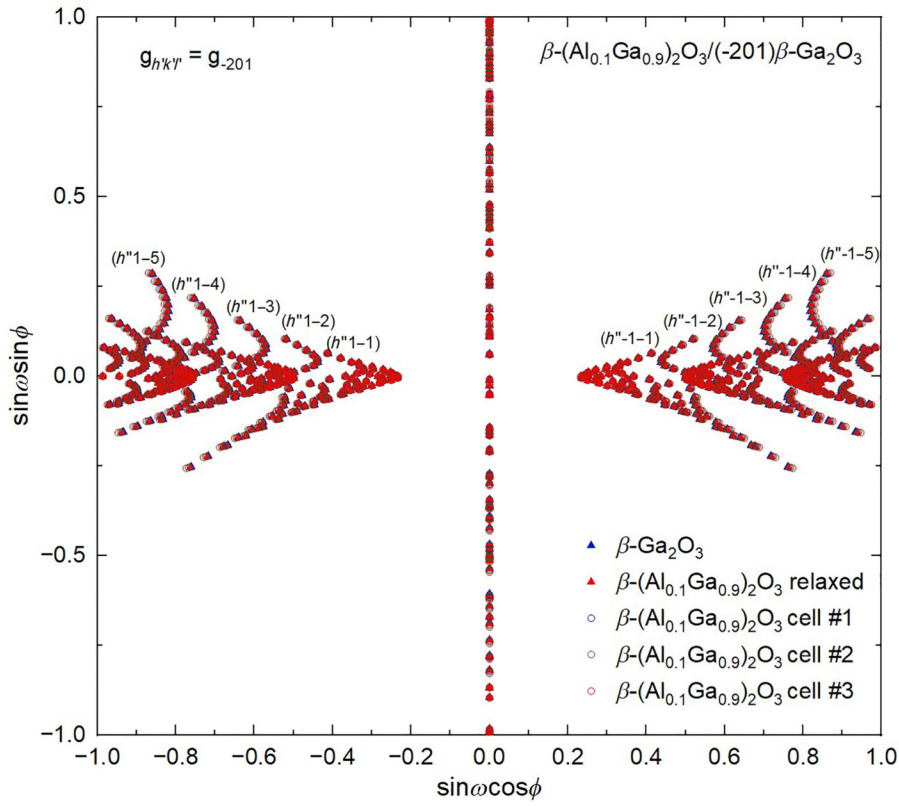


FIG. 12. Same as Fig. 10 for $\mathbf{g}_{h'k'l'} = \mathbf{g}_{\bar{2}01}$.

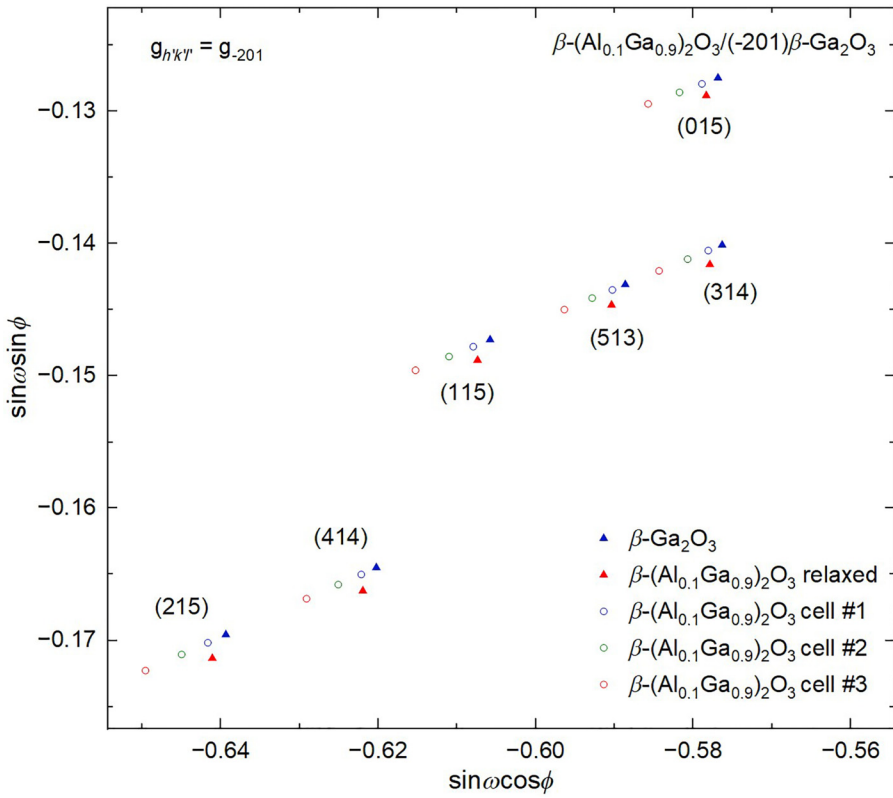


FIG. 13. Detail for Fig. 12 with $\mathbf{g}_{h'k'l'} = \mathbf{g}_{\bar{2}01}$.

with the diffraction peaks from the substrate, while the diffraction peak from the strain-free epitaxial layer with the same aluminum content is shifted along different directions. The appearance of the three strained unit-cell lattice diffraction peaks along lines sharing the diffraction peak from the strain-free substrate is caused by the fact that all three strained unit cells here share one common plane, $(\bar{2}01)$, with the substrate. Accordingly, there is a coordinate direction within the Ewald sphere along which all pseudomorphically grown epitaxial layers will follow depending on their state of strain and composition and which will include the same lattice diffraction peak from the substrate. A more qualitative and quantitative assessment of these coordinate directions, which also depend on the choice of the lattice plane (h', k', l') , will be discussed elsewhere. The latter statement can be seen when interrogating Fig. 12. Here, we display the same diffraction peak conditions but for $(h'k'l') = (\bar{2}01)$. As may be expected, due to the higher symmetry of this plane compared to $(\bar{2}\bar{2}2)$ in Fig. 10, the overall diffraction pattern reveals the monoclinic mirror symmetry about the plane **(a, c)**, which is vertical to the figure plane, and at $\phi = 0, \pm\pi$. The axis **b** is parallel to $\mathbf{u}_2$. Diffraction peaks at $\phi = 0, \pm\pi$ are due to the planes (h'', k'', l'') with $k'' = 0$. Selected series of diffraction peaks are indicated, for example, $(h'', 1, \bar{5})$, $(h'', 1, \bar{4})$, $(h'', 1, \bar{3})$, $(h'', 1, \bar{2})$, $(h'', 1, \bar{1})$, and the mirror-symmetric series $(h'', \bar{1}, 1)$, $(h'', \bar{1}, 2)$, $(h'', \bar{1}, 3)$, $(h'', \bar{1}, 4)$, and $(h'', \bar{1}, 5)$. Figure 13 depicts a detail from Fig. 12. Here, similar shifts along straight lines of the strained unit-cell diffraction peaks away from the corresponding strain-free substrate diffraction peaks can be seen. Likewise, the diffraction peak from the strain-free epitaxial layer is shifted to locations not coinciding with the lines formed by the diffraction peaks of the pseudomorphic epitaxial layers. These diagrams, shown in Figs. 10–13, may serve as maps to search for the occurrence of the differently strained unit-cell solutions in experimental data of epitaxial layers grown on $(\bar{2}01)$ β -Ga₂O₃, as predicted in this work. The algorithm discussed above and the equations for computation of the unit-cell solutions can be used to predict any x-ray diffraction diagram for any other surface orientation, scattering condition, and strain condition due to pseudomorphic growth on any surface $(h0l)$.

VI. CONCLUSIONS

The approach introduced here reveals the existence of exactly three different unit cells under a given biaxial strain situation for strain in pseudomorphically grown heteroepitaxial systems with low crystal symmetries. In this work, we demonstrated the approach for monoclinic symmetry and a special class of surfaces, $(h0l)$. In this example, the symmetry of the elastically stable epitaxial layer remains monoclinic. We find that for almost all surfaces in this class of templates, two domains can exist whose stress-free

directions are not perpendicular to the surface; however, one domain is favored both energetically and by having its stress-free direction closer to the surface normal than the others. The approach presented here is generally applicable to higher symmetries, and by further modification also to triclinic materials. It is anticipated that in cases for which the template surface has none of the lattice unit-cell axes aligned within its plane, the biaxial strain will reduce the symmetry of the monoclinic epitaxial layer to triclinic and introduce shear strains and shear stresses in all elements of ϵ and σ . Then, all three unit cells may compete in epitaxial growth with stress-free directions neither parallel nor perpendicular to the template surface. The effects of the differently strained unit-cell solutions are demonstrated by calculating the strain-induced shifts of the three lowest electronic band-to-band transition energies in β -(Al_xGa_{1-x})₂O₃ for $x = 0.1$ grown pseudomorphically onto β -Ga₂O₃ templates.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- [58] Note the structure of the determinant for the stress tensor, where the component σ_{zz} always factorizes out. This yields

trivial solutions for all cases when the b axis is perpendicular to the surface, (010). Then the z direction is also parallel to the surface normal $\mathbf{g}_{hkl}$. Hence, for growth on (010), only one stress-free solution exists, with a stress-free direction not parallel to the surface, which is then obviously the surface normal. For almost all other crystallographic surfaces, the stress-free directions are neither parallel nor perpendicular to the surface, and all represent potential stable unit cells during growth.