On the applicability of elastic model to very thin crystalline layers

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On the applicability of elastic model to very thin crystalline layers

G Jurczak[1,3]**, M Maździarz**[1]**, P Dłużewski**[1]**, G P Dimitrakopulos**[2]**, Ph Komninou**[2]**, and T Karakostas**[2]

[1] Institute of Fundamental Technological Research, Polish Academy of Sciences, ul. Pawińskiego 5b, 02-106 Warsaw, Poland
[2] Department of Physics, Aristotle University of Thessaloniki, GR 54124 Thessaloniki, Greece

Abstract. Elastic model of continuum material is often used to simulate the relaxation of crystalline heterostructures. There are many reports on the successful application of the theory of elasticity to nano-sized crystalline heterostructures, even if the continuum condition for them is hardly fulfilled. On the other hand, progress in epitaxial growth allows for the preparation of stable ultra-thin layers with thickness of few monolayers. For such ultra-thin layers, results provided by continuum model and molecular statics/dynamics calculations become diverging. The key problem seems to be located at the modelling of the interface between layers, which is problematic in the continuum approach. By applying a step-wise substitutive compositional interfacial function, it is possible to obtain good agreement with molecular dynamics calculations, even for a single monolayer heterostructure. We propose another approach that uses composition as an extra parameter during finite element calculations, along with classical nodal displacements. Such an approach creates a chemo-elastic coupling that allows to interpolate the composition much like in the case of atomistic calculations.

1. Introduction

In recent decades, III-nitride heterostructures in the form of quantum wells (QW), wires (QWR), and dots (QD) found many applications in optoelectronics, e.g. they have been used as structural elements in light-emitting and laser diodes, sensors, photovoltaic devices, etc. To enhance the intensity of light emission and other special capabilities of these nano-heterostructures, e.g. in the long wavelength operational range [1,2], a high indium content in ternary and quaternary III-N semiconductors is exploited. Unfortunately, the behaviour of the indium element, lack of suitable templates, and large lattice mismatch cause indium clustering and high densities of extended defects [3,4,5,6]. A solution of indium alloying problems is expected by the use of very thin, binary InN layers in the form of QWs. These thin layers, with thicknesses below critical, are arranged in short period superlattices (SPS) [4,7].

The composition of a crystalline heterostructure, as well as the lattice strain, which are related to each other can be estimated by precise measurements of the positions and intensities of atomic columns using high resolution (scanning) transmission electron microscopy (HR(S)TEM) [7,8]. In order to establish a connection between strain and composition, a full elastic accommodation of the misfit and Vegard's law are usually assumed. To describe relaxation of a crystalline heterostructure, the theory of elasticity is employed, which uses a continuum model of the material. There are many reports on the successful application of elastic model to nano-sized crystalline heterostructures (see for example [8,9,10,11]). However, for ultra-thin layers, elasticity does not accurately describe the

relaxation of the heterostructure. The results provided by elasticity differ from molecular statics/dynamics calculations. The problem seems to be addressed to the interfaces between atomic layers, and requires a distinct approach pertaining to the chemical composition. To reproduce atomistic composition at the interface, it is necessary to introduce a substitutive compositional function in the continuum model of the interface. This is crucial in the case of very thin layers, with thickness of few monolayers (MLs), where the interfaces constitute large part of the layer.

In this paper, the elastic relaxation of binary and ternary III-N heterostructures in the form of 1 ML and 2 MLs QWs are considered by use of elasticity and molecular dynamics (MD). A modified continuous approach with the compositional interfacial function x within the interface allows to take into account the specific character of very thin layers and to obtain results in agreement with molecular dynamics calculations.

2. Computational scheme
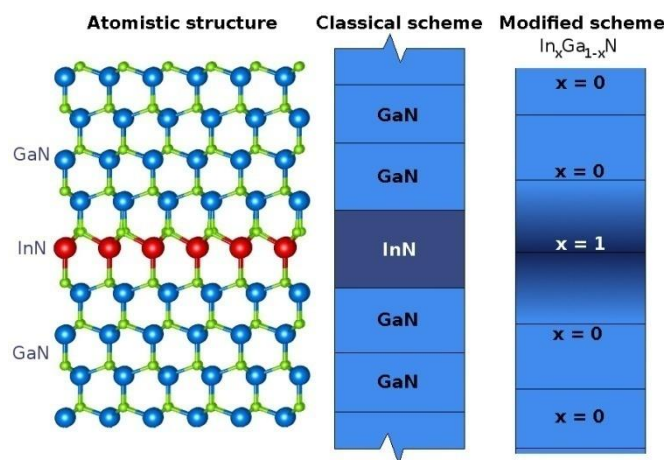
Figure 1. Atomistic, classical continuous, and modified continuous (with additional, compositional interfacial function x) schemes of the 1 ML InN/GaN hetero-structure.

Let us consider $In_{0.33}Ga_{0.67}N$/GaN and InN/GaN, polar heterostructures which consist of 5 QWs with nominal thickness equal to 1 ML and 2 MLs, as described in [7]. The thickness of the GaN barriers was equal to 10 lattice parameters c. The bottom and top GaN layers of our computational samples were limited to $20c$. Since our heterostructure has in-plane dimensions much larger than its thickness, it is justified to limit the in-plane size of our sample and apply Dirichlet-type boundary conditions (BCs) instead. As a result, elastic relaxation is allowed only along the growth direction, i.e. along c-axis. To determine the displacement and strain fields of the relaxed heterostructure, a boundary value problem of the elasticity has to be formulated and solved either analytically or numerically, for example by the finite element method (FEM). The properties of ternary alloys were calculated by use of Vegard's law and elastic parameters of the binary crystals given in [12]. FEM calculations were
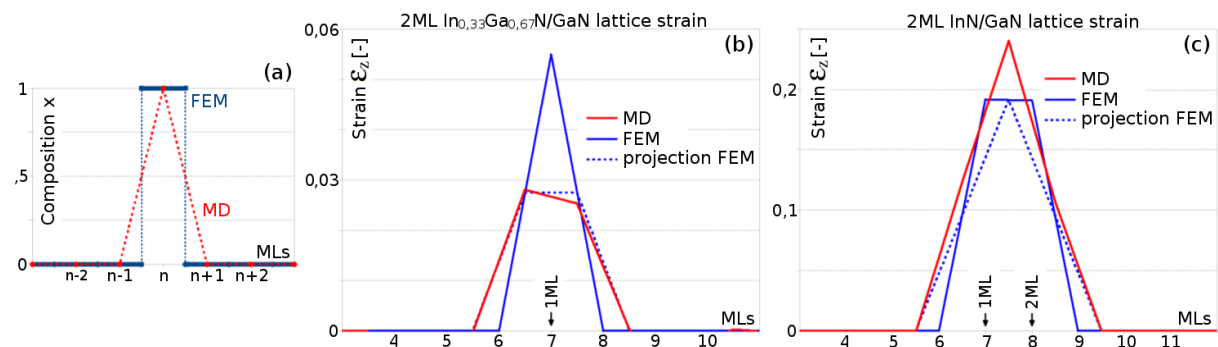
Figure 2. Compositional changes for 1 ML InN/GaN heterostructure (a), and lattice strains in atomistic and classical continuum approach: b) for 1 ML of $In_{0.33}Ga_{0.67}N$/GaN and c) for 2 MLs of InN/GaN heterostructure. Dashed lines in compositional chart denote assumed approximation for substitutive compositional function x at the interface. Charts show distribution for a single QW only.

performed in the FEAP program [13] with use of 3D 8-node solid elements [14]. The FE mesh coincides with atomic layers to be consistent with MD calculations, since d-spacing strain is calculated on the basis of average displacements between MLs. In the case of MD simulations, the supercells were relaxed by use of a bond-order, many body Tersoff potential in LAMMPS software [15]. More details concerning atomic interactions and the parameterization are given in [7]. Atomistic and continuous models of the 1 ML InN/GaN heterostructure are depicted in Figure 1. The local coordinate system **XYZ** is assumed that way **Z** axis corresponds to the growth direction. According to the applied BCs and relaxation scheme, the shear stresses vanish, while normal stresses are (Voight notation):

$$\sigma_x = C_{11}\varepsilon_x^e + C_{12}\varepsilon_y^e + C_{13}\varepsilon_z^e \neq 0,$$
$$\sigma_y = C_{12}\varepsilon_x^e + C_{11}\varepsilon_y^e + C_{13}\varepsilon_z^e \neq 0, \qquad (1)$$
$$\sigma_z = C_{13}\varepsilon_x^e + C_{13}\varepsilon_y^e + C_{33}\varepsilon_z^e = 0,$$

where C_{ij} means stiffness coefficients. In our model, the lattice (total) strain $\boldsymbol{\varepsilon}$ is the sum of elastic $\boldsymbol{\varepsilon}^e$ and chemical (lattice mismatch) $\boldsymbol{\varepsilon}^{ch}$ strains assuring consistency with HR(S)TEM strain measurements [7,8]. Lack of lattice in-plane strains $\varepsilon_x = \varepsilon_y = 0$ comes from the Dirichlet-type BCs, so elastic in-plane strains are:

$$\varepsilon_x^e = -\varepsilon_x^{ch} = \varepsilon_y^e = -\varepsilon_y^{ch} = -\left(\frac{a^{QW}}{a^M} - 1\right), \qquad (2)$$

where a^{QW} is an appropriate lattice parameter of the $In_{0.33}Ga_{0.67}N$ or InN QW layer, and a^M is the lattice parameter of the GaN barrier. Finally, we may calculate the ε_z^e strain as:

$$\varepsilon_z^e = \frac{2C_{13}}{C_{33}}\varepsilon_x^{ch}. \qquad (3)$$

By substituting the right lattice parameters to Equation [2] and elastic stiffness coefficients to Equation [3], the expected elastic strains ε_z^e in our heterostructures are equal to 2.19% for $In_{0.33}Ga_{0.67}N$ and 9.17% for InN layer. Lattice mismatches ε_z^{ch}, related to c-axis relaxation direction, according to Equation [2] are equal to 3.30% and 9.99%, respectively. To sum up, lattice strains ε_z are equal to 5.49% and 19.16%, respectively. These values correspond to d-spacing strain calculated based on FE mesh displacements, see Figure 2. Figure 2 shows also the d-spacing strain distribution calculated by MD [7] and nodal projections of FE strains, i.e. displacements of the nodes $\boldsymbol{u}$ multiplied by derivatives of the shape functions $\nabla\mathbf{N}$, analogously to small strain tensor [13]:

$$\boldsymbol{\varepsilon}^e = \frac{1}{2}(\nabla^T\mathbf{u} + \nabla\mathbf{u}). \qquad (4)$$

Then, elastic strain calculated in Gauss points must be projected back to nodes with a proper weight. d-spacing strain distributions and its values obtained by continuum (FEM) and atomistic approach (MD) differ significantly, see for example 1 ML QW in Figure 2(b). In the case of 2 MLs, the d-spacing strain discrepancy is much smaller, but the strain distribution is still different. The FE nodal projection of the lattice strain repeats the MD d-spacing curves and gives results far better than d-spacing strain calculated based on nodal displacements. Anyway, displacements of nodes contain important information about the relaxation of the sample, for example in the case of reconstruction of the actual atomic structure. Therefore, modification of the classical elastic approach is necessary to get agreement with MD calculations. Modifications in strain distribution done by nodal projection suggests that any heterostructure interface should be modelled by applying a substitutive composition
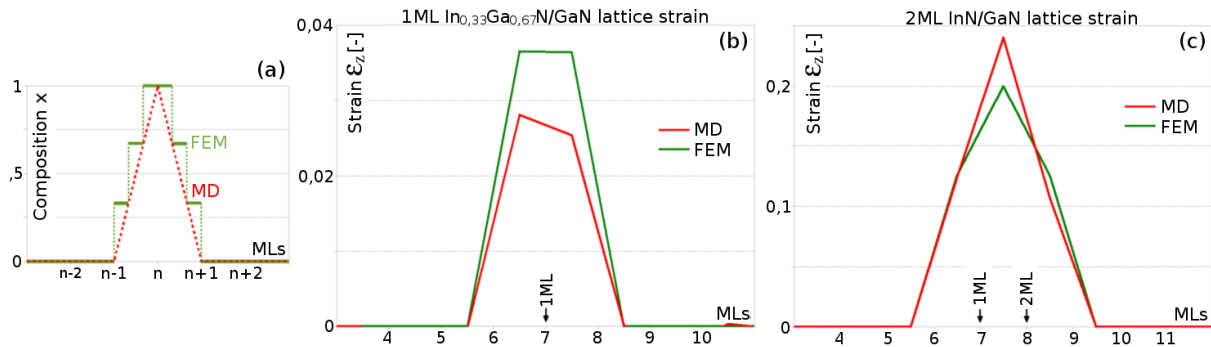
Figure 3. The same as in Figure 2 but now for modified elastic approach (additional sub-elements at the interface).

function, much like in the case of interpolation of atomistic composition, see Figure 2(a). The classical continuum approach assumes rectangular, sharp interfaces with an abrupt change of composition and physical properties. Distinct interpretation of the interface by the continuum and atomistic approach results in different *d*-spacing strains and their distributions.

To reduce the discrepancy in interpretation of the interface composition function, let us modify the FE mesh by use of extra sub-layers corresponding to compositional grading, as depicted in Figure 3(a). The accuracy of this approach depends on the quality of the interpolation, i.e. on the number of sub-layers. The drawback of the approach is a procedural one, pertaining to a step-like interpolation of a compositional function. The more steps, the more precise is the interpolation. Unfortunately, this also increases the number of sub-elements at the interface and the work quantity to prepare calculations in each sub-element. Results presented in Figure 3 seems to be somewhat overestimated. It is an effect of the assumed compositional interpolation based on 3 sub-layers at the interface area with step-like composition interpolation. Assumed composition function overestimates the indium content in comparison to the atomistic calculations. Thereby, *d*-spacing strains related to a continuum model overestimate atomistic results as well.

To avoid the drawback of the approach based on interfacial sub-layers, we propose a new methodology within the framework of continuum approach, which uses additional, compositional parameter at the FE node (addition to classical displacements). Using FE shape functions [14] it is possible to span the composition over the element and get continuum interfacial composition function *x*. Composition function modify the right-hand side vector of the global matrix equation solved by FEM [13]:

$$[K][q] = [Q],$$
$$Q_i = \sum_{l=1}^{M}\left[C_{ij}\,\varepsilon_j^{ch}(x_l)\,\nabla_i N_l\right]\Delta V_l. \tag{5}$$

Square matrix K_{ij} denotes the so-called stiffness matrix, q_j is the vector of nodal degrees of freedom, and the right-hand side vector Q_i is related to external forces acting in nodes. The local mismatch strain $\varepsilon^{ch}(x_l)$ is related to composition x_l interpolated at a given Gauss point l. ∇N denotes the derivative of the element's shape function, while ΔV is a weight function related to element volume. *d*-spacing strains depicted in Figure 4 were calculated with use of a compositional parameter at FE nodes. The assumed substitutive composition repeats an atomistic layout presented in Figure 2.
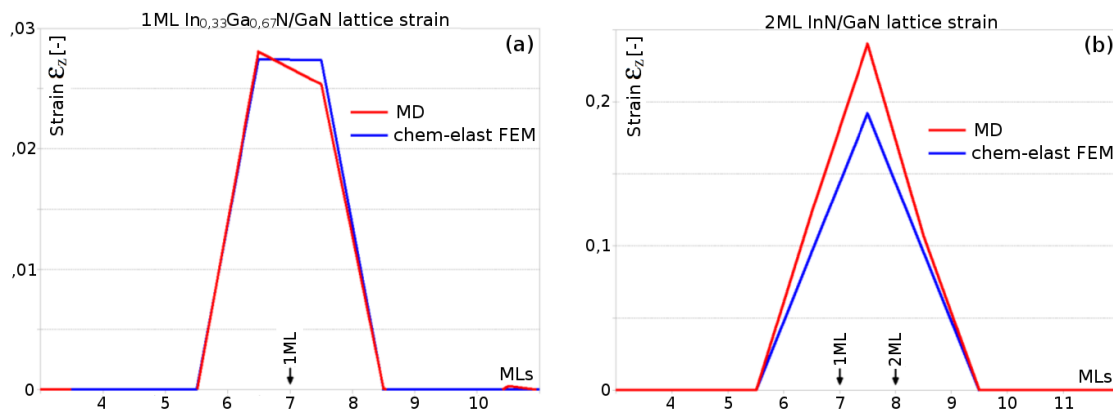
Figure 4. Lattice strains calculated by atomistic and chemo-elastic approach: a) for 1ML of $In_{0.33}Ga_{0.67}N$ /GaN and b) for 2MLs of InN/GaN hetero-structure. Composition function of chemo-elastic approach reproduces an atomistic model, see Figure 2(a).

3. Summary
Ultra-thin crystalline heterostructures need more attention during modelling of their elastic relaxation due to the interfacial influences. Abrupt compositional changes at the interface in the case of a classical elastic approach are not in line with atomistic modelling. To avoid such discrepancies within a classical, elastic approach, extra sub-layers may be introduced at the interfacial region with appropriate step-like composition and elastic properties. Another option is to apply an additional nodal

parameter related to a layer composition. In this way it is possible to get consistent displacement and *d*-spacing strain fields in the case of continuum and atomistic modelling of the elastic relaxation. The compositional interface function seems to be more attractive approach compared to the sub-layers approach as it requires only small changes in the FEM code and creates more versatile chemo-elastic coupled problem.

Acknowledgments

References

[1]   Chichibu S, Azuhata T, Sota T and Nakamura S 1997 *Appl. Phys. Lett.* **70** 2822
[2]   Pan W, Dimakis E, Wang G T, Moustakas T D and Tsui D C 2014 *Appl. Phys. Lett.* **105** 213503
[3]   ODonnell K P, Mosselmans J F W, Martin R W, Pereira S and White M E 2001 *J. Phys. : Condens. Matter* **13** 6977
[4]   Morales F, Gonzalez D, Lozano J G, Garcia R, Hauguth-Frank S, Lebedev V, Cimalla V and Ambacher O 2009 *Acta Materialia* **57** 5681
[5]   Gorczyca I, Łepkowski S P, Suski T, Christensen N E and Svane A 2009 *Phys. Rev. B* **80** 075202
[6]   Bazioti C, Papadomanolaki E, Kehagias T, Walther T, Smalc-Koziorowska J, Pavlidou E, Komninou P, Karakostas T, Iliopoulos E and Dimitrakopulos G P 2015 *J. Appl. Phys.* **118** 155301
[7]   Dimitrakopulos G P, Vasileiadis I G, Bazioti C, Smalc-Koziorowska J, Kret S, Dimakis E, Florini N, Kehagias T, Suski T, Karakostas T, Moustakas T D and Komninou P 2018 *J. Appl. Phys.* **123** 024304
[8]   Dimitrakopulos G P, Kalesaki E, Kioseoglou J, Kehagias T, Lotsari A, Lahourcade L, Monroy E, Husler I, Kirmse H, Neumann W, Jurczak G, Young T D, Dłużewski P, Komninou P and Karakostas T 2010 *J. Appl. Phys.* **108** 104304
[9]   Dłużewski P, Maciejewski G, Jurczak G, Kret S and Laval J Y 2004 *Comput. Mater. Sci.* **29** 379
[10]   Hol V, Springholz G, Pinczolits M and Bauer G 1999 *Phys. Rev. Lett.* **83** 356
[11]   Liu Y and Ye W 2017 Physica E **89** 5
[12]   Vurgaftman I and Meyer J R 2003 *J. Appl. Phys.* **94** 3675
[13]   Taylor R L 2003 *FEAP - A Finite Element Analysis Program, Theory Manual* (University of California, Berkley) www.ce.berkeley.edu/projects/feap/
[14]   Maździarz M 2010 *CMES: Computer Modeling in Engineering & Sciences* **66**(1) 1-24
[15]   Plimpton S 1995 *J. Comp. Phys.* **117** 1 http://lammps.sandia.gov