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Static and dynamic effects due to electrostriction in GaN/AlN

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Abstract
A one-dimensional model accounting for electrostriction, lattice mismatch, piezoelectricity, and strain is presented with special emphasis on GaN/AlN heterostructures recently examined extensively in the literature. It is shown that electrostriction, being a second-order effect in the strain–electric field relation, plays a significant, sometimes dominant, contribution subject to DC voltage conditions and externally imposed hydrostatic pressure or AC conditions. It is argued that electrostriction, examined in a three-dimensional model, may give important contributions to the total strain at DC conditions even in the absence of hydrostatic pressure. Model results are based on experimentally reported values for electrostriction coefficients in GaN.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Wurtzite heterostructure material combinations based on nitrides are promising candidates for optical and electronic device applications. Several detailed computational methods for analysing the electronic properties of strained semiconductor heterojunctions have been reported recently (e.g. [1–6]). However, it is still unclear which are the most significant strain contributions in nanostructures. Due to large electric fields (of the order of several hundreds of MV m^{-1}) in nanostructures [1, 4, 5], it is important to assess whether higher-order strain–electric field effects play a significant role in addition to piezoelectric effects, nonlinearities in the permittivity, and electric charge screening. Recently, nonlinear piezoelectricity has been proposed as being important in InGaN nanostructures [7]. In the present work, the second-order electrostriction effect existing in all semiconductor materials (electrostriction does not require inversion asymmetry in the unit cell) is examined based on a one-dimensional model and shown to give a significant (sometimes dominant) strain contribution when (a) DC voltage conditions are imposed (exemplified by short-circuiting a GaN/AlN heterostructure) in addition to hydrostatic pressure, or (b) under AC conditions without externally imposing a hydrostatic pressure. Measurements of the electrostriction constant in GaN [8] indeed reveal that electrostriction coefficients are one to two orders of magnitudes larger than in PVDF [9] where they are known to be important. This coefficient has also been recently used in a phenomenological model for the determination of the spontaneous polarization [10]. Moreover, electrostriction may lead to significant strain effects even for DC conditions without imposing hydrostatic pressure in a three-dimensional analysis.

2. Theory

The governing equations for a lattice-mismatched heterostructure consisting of two wurtzite materials (GaN and AlN) accounting for piezoelectric effects and spontaneous polarization read

$$\mathbf{T} = \mathbf{cS} - \mathbf{eE}, \tag{1}$$

$$\mathbf{D} = \boldsymbol{\epsilon}\mathbf{E} + \mathbf{P}^{\mathrm{spon}} + \mathbf{eS} + \mathbf{MTE}, \tag{2}$$

$$\nabla \cdot \mathbf{T} = \rho_{\mathrm{d}} \frac{\partial^2 \mathbf{u}}{\partial t^2}, \tag{3}$$

$$\nabla \cdot \mathbf{D} = \rho_{\mathrm{f}}, \tag{4}$$

where $\mathbf{T}, \mathbf{S}, \mathbf{D}, \mathbf{E}, \mathbf{P}^{\mathrm{spon}}$, and $\mathbf{u} = (u_x, u_y, u_z)$ are the stress tensor, strain tensor, electric displacement, electric field, spontaneous polarization, and the particle displacement, respectively. The coefficients $\mathbf{c}, \boldsymbol{\epsilon}, \mathbf{e}, \mathbf{M}, \rho_{\mathrm{d}}$, and ρ_{f} denote the position-dependent elastic tensor, permittivity tensor, piezoelectric $\mathbf{e}$ tensor, electrostriction coefficient, mass density, and the free-charge carrier density, respectively. Note that the electrostrictive term in equation (2) is the so-called converse electrostrictive effect. To simplify the description of electrostriction (and because limited experimental information is available for electrostrictive coefficients) we shall consider only the z-component part: $M_{zzzz} T_{zz} E_z$ [8] in the following (i.e. an electrostrictive contribution exists only for D_z). Furthermore, the strain $\mathbf{S}$ accounts for lattice mismatch as well as the direct electrostriction effect and is defined by:

$$\begin{aligned}
S_1 &= \frac{\partial u_x}{\partial x} - \frac{a(\vec{r}) - a^1}{a^1}, & S_4 &= \frac{1}{2}\left(\frac{\partial u_y}{\partial z} + \frac{\partial u_z}{\partial y}\right), \\
S_2 &= \frac{\partial u_y}{\partial y} - \frac{a(\vec{r}) - a^1}{a^1}, & S_5 &= \frac{1}{2}\left(\frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x}\right), \\
S_3 &= \frac{\partial u_z}{\partial z} - \frac{c(\vec{r}) - c^1}{c^1} + M_{zzzz} E_z^2, & S_6 &= \frac{1}{2}\left(\frac{\partial u_x}{\partial y} + \frac{\partial u_y}{\partial x}\right),
\end{aligned} \tag{5}$$

where $a(\vec{r}), c(\vec{r})$ are the lattice constants at position $\vec{r}$ while a^1, c^1 denote the lattice constants of the substrate. Again, we choose to consider only the z-component part of the direct electrostrictive effect in the strain tensor. The same coefficient M_{zzzz} appears in the expressions for D_z and S_3 for the electrostrictive contribution, as can be proved using thermodynamic considerations based on Gibbs' free energy.

2.1. DC conditions subject to hydrostatic pressure

It is evident from the governing equations (1)–(4) that electrostriction will be effective if hydrostatic pressure is present in the system considered. It turns out, as we shall see, that the influence of electrostriction in a one-dimensional model for the total strain and electric field under static conditions disappears unless stress is present. Hence, we will assume that a

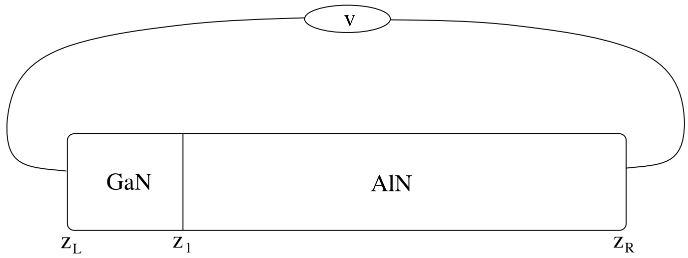

Figure 1. Plot of the heterostructure consisting of a GaN film layer deposited on an AlN substrate.

hydrostatic pressure is imposed on the two-layer GaN/AlN heterostructure shown in figure 1. The more compact notation $M_{33} = M_{zzzz}$, $E_3 = E_z$, and $D_3 = D_z$ is used in the following.

Under static conditions, the z-component equation of motion—with continuity in stress throughout the structure—immediately gives that the stress component T_3 is constant in the structure, say T_0, due to the imposed hydrostatic pressure. Hence, we have

$$c_{33}\frac{\partial S_3}{\partial z} - e_{33}\frac{\partial E_3}{\partial z} = 0, \tag{6}$$

$$T_0 = c_{13}S_1 + c_{13}S_2 + c_{33}S_3 - e_{33}E_3. \tag{7}$$

Moreover, Maxwell's equation (equation (4)) yields

$$\frac{\partial D_3}{\partial z} = \epsilon_{33}\frac{\partial E_3}{\partial z} + e_{33}\frac{\partial S_3}{\partial z} + M_{33}T_0\frac{\partial E_3}{\partial z} = 0. \tag{8}$$

Combining equations (6) and (8) immediately shows that the electric field and strain is constant in each layer (but not continuous across layers!). With this knowledge, we can assign E^{A}, S^{A} and E^{B}, S^{B} as the electric field, strain component 3 in layer A (GaN) and layer B (AlN), respectively. Since the lattice mismatch is constant and different from zero only in layer A, we can now write down two equations in the four coefficients E^{A}, S^{A}, E^{B}, S^{B} by imposing equation (7) and continuity of stress:

$$T_0 = -2c_{13}^{\mathrm{A}}\frac{a^2 - a^1}{a^1} + c_{33}^{\mathrm{A}}S^{\mathrm{A}} - e_{33}^{\mathrm{A}}E^{\mathrm{A}}, \tag{9}$$

$$T_0 = c_{33}^{\mathrm{B}}S^{\mathrm{B}} - e_{33}^{\mathrm{B}}E^{\mathrm{B}}, \tag{10}$$

where superscripts on parameters denote their values in the appropriate layer and a^2 is the lattice constant of GaN parallel to the GaN/AlN interface. Assuming electrical short-circuiting the full structure gives:

$$E^{\mathrm{A}}(z_1 - z_{\mathrm{L}}) + E^{\mathrm{B}}(z_{\mathrm{R}} - z_1) = 0. \tag{11}$$

The remaining equation needed to determine the electric field and the strain values in each layer comes from imposing continuity in the normal electric displacement component across the GaN/AlN interface, i.e.

$$\epsilon_{33}^{\mathrm{A}}E^{\mathrm{A}} + P_{\mathrm{spon}}^{\mathrm{A}} - 2e_{31}^{\mathrm{A}}\frac{a^2 - a^1}{a^1} + e_{33}^{\mathrm{A}}S^{\mathrm{A}} + M_{33}^{\mathrm{A}}T_0E^{\mathrm{A}} - e_0 n$$
$$= \epsilon_{33}^{\mathrm{B}}E^{\mathrm{B}} + P_{\mathrm{spon}}^{\mathrm{B}} + e_{33}^{\mathrm{B}}S^{\mathrm{B}} + M_{33}^{\mathrm{B}}T_0E^{\mathrm{B}}, \tag{12}$$

where e_0 is the free-electron charge. In deriving equation (12), an interface charge density $n\delta(z - z_1)$ at the GaN/AlN interface has been included following [1] as it is known to lead to compensation of otherwise too large electric fields in GaN/AlGaN heterostructures. The solution of the four equations above determines the four coefficients E^{A}, S^{A}, E^{B}, S^{B}.

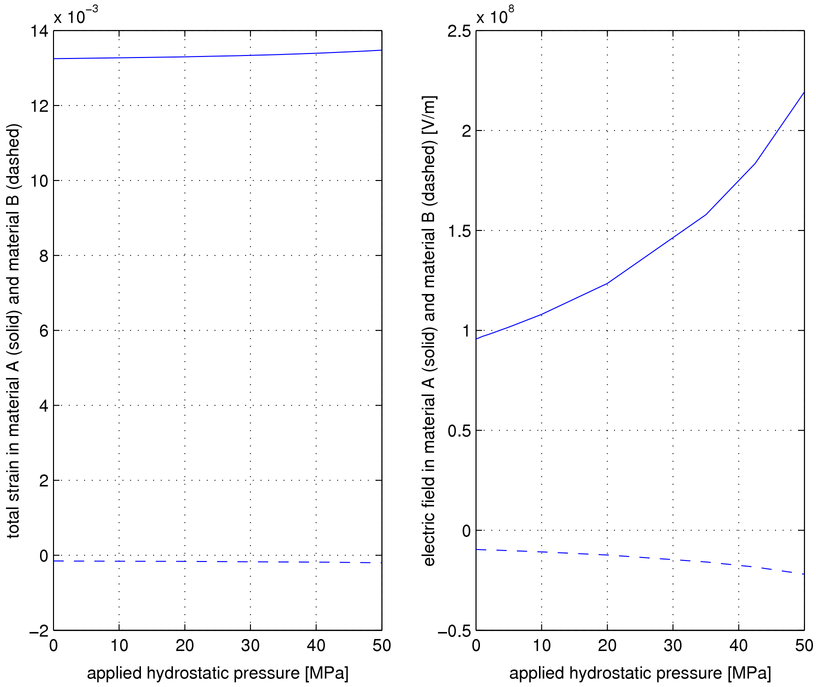

Figure 2. Left plot: total strain values in the GaN film layer (solid) and the AlN substrate (dashed) as a function of the externally applied hydrostatic pressure. Right plot: electric-field value in the GaN film layer (solid) and the AlN substrate (dashed) as a function of the externally applied hydrostatic pressure. The heterostructure is short-circuited and the GaN layer thickness is 10 nm.

Evidently, the influence of electrostriction in a one-dimensional model is possible only if $T_0 \neq 0$ subject to DC conditions. However, electrostriction gives an (important) effect in GaN/AlN if a hydrostatic pressure is present in the system, as we shall see next. A hydrostatic pressure of $T_0 = 0.1$ GPa [11], an electric field of approximately $E^{\mathrm{A}} = 1 \times 10^8$ V m^{-1}, and $M_{33} = 1 \times 10^{-18}$ m^2 V^{-2} gives a contribution to the electric displacement of approximately 0.01 C m^{-2} comparable to the usually strongest contribution: $\epsilon_{33}^{\mathrm{A}} E^{\mathrm{A}} = 0.008$ C m^{-2}. Note that a three-dimensional DC analysis of electrostriction will not require an externally imposed hydrostatic pressure in order to have a strong influence of electrostriction.

In figure 2 (left), plots of the total strain values S^{A} and S^{B} in the GaN and AlN layers, respectively, are shown versus the external hydrostatic pressure for a GaN layer thickness of 10 nm. Notice that the total strain in the GaN layer increases slightly from 1.32% to 1.34% as the hydrostatic pressure increases from 0 to 50 MPa, while the total strain in the AlN layer is basically unaffected by the externally applied hydrostatic pressure (remains nearly constant at a value of −0.015 %). The electric field in the GaN layer increases markedly, however, from nearly 1×10^8 V m^{-1} to above 2×10^8 V m^{-1} as the hydrostatic pressure increases from 0 to 50 MPa (figure 2 (right)). Similarly, the electric field in the substrate increases (in absolute values) with increasing hydrostatic pressure from approximately 0.1×10^8 to above 0.2×10^8 V m^{-1} as the hydrostatic pressure increases from 0 to 50 MPa.

Electric-field results in GaN structures subject to hydrostatic pressure have been reported experimentally on more complex multi-quantum-well structures than the simple structure considered here, leading to (in [12]) electric-field values close to or above electric breakdown. For these reasons, we have not made a comparison with the results of [12]. It should also be noted that the stiffness coefficients of GaN and AlN are functions of hydrostatic pressure as well. Recent work by Lepkowski *et al* [13, 14] indicates, however, that changes in the stiffness tensor due to externally applied hydrostatic pressures are very small (less than approximately 0.5% as the hydrostatic pressure increases from 0 to 10 kbar) and hence can be neglected in comparison with the strong electrostrictive effect found in the present work.

2.2. Constant electric displacement in space. Monofrequency time dependence

Next, we consider the case where a small AC electric field is superimposed upon a (larger) DC electric field in a single layer of GaN for the purpose of examining resonance characteristics in a linear AC model approach. In this case, electrostriction plays a strong role for the values of resonance frequencies even in the absence of an externally applied hydrostatic pressure, as we shall see next. In the following, all AC quantities are denoted with a tilde. Since the spontaneous polarization and the lattice-mismatch part of the strain are independent of time, we have from equation (2)

$$\tilde{D}_3 = \epsilon_{33}\tilde{E}_3 + e_{33}\tilde{S}_3 + M_{33}E_3^0\tilde{T}_3, \tag{13}$$

as the DC stress is zero. Furthermore

$$\tilde{T}_3 = c_{33}\tilde{S}_3 - e_{33}\tilde{E}_3, \tag{14}$$

which by insertion in equation (13) yields

$$\tilde{E}_3 = \frac{1}{\epsilon_{33} - M_{33}E_3^0 e_{33}}\tilde{D}_3 - \frac{e_{33} + M_{33}E_3^0 c_{33}}{\epsilon_{33} - M_{33}E_3^0 e_{33}}\tilde{S}_3 \equiv \frac{1}{\epsilon'_{33}}\tilde{D}_3 - \frac{e'_{33}}{\epsilon'_{33}}\tilde{S}_3, \tag{15}$$

and the last equality defines the coefficients ϵ'_{33} and e'_{33}. Navier's equation for the AC part of T_{33} (equation (3)) gives

$$\frac{\partial \tilde{T}_3}{\partial z} = \rho_\mathrm{d}\frac{\partial \tilde{v}_3}{\partial t}, \tag{16}$$

or from equation (14) assuming monofrequency conditions

$$c_{33}\frac{\partial \tilde{S}_3}{\partial z} - e_{33}\frac{\partial \tilde{E}_3}{\partial z} = \mathrm{i}\omega\rho_\mathrm{d}\tilde{v}_3. \tag{17}$$

The strain equation for $\tilde{S}_3$ reads

$$\frac{\partial \tilde{S}_3}{\partial t} = \frac{\partial \tilde{v}_3}{\partial z} + 2M_{33}E_3^0\frac{\partial \tilde{E}_3}{\partial t}, \tag{18}$$

i.e.

$$\mathrm{i}\omega\tilde{S}_3 = \frac{\partial \tilde{v}_3}{\partial z} + 2\mathrm{i}\omega M_{33}E_3^0\tilde{E}_3. \tag{19}$$

Differentiating equation (17) with respect to z and using equations (15) and (19) allows us to augment $\tilde{E}_3$, $\tilde{v}_3$ so as to obtain a single ordinary differential equation for $\tilde{S}_3$:

$$\left(c_{33} + \frac{e_{33}e'_{33}}{\epsilon'_{33}}\right)\frac{\partial^2 \tilde{S}_3}{\partial z^2} + \omega^2\rho_\mathrm{d}\left[1 + 2M_{33}E_3^0\frac{e'_{33}}{\epsilon'_{33}}\right]\tilde{S}_3 = 2\omega^2\rho_\mathrm{d}\frac{M_{33}E_3^0}{\epsilon'_{33}}\tilde{D}_3. \tag{20}$$

The solution to the above equation $\tilde{S}_3$ generally reads

$$\tilde{S}_3(z) = A\exp(\mathrm{i}kz) + B\exp(-\mathrm{i}kz) + \frac{2M_{33}E_3^0}{\epsilon'_{33} + 2M_{33}E_3^0 e'_{33}}\tilde{D}_3, \tag{21}$$

$$k = \sqrt{\rho_\mathrm{d}\frac{\epsilon'_{33} + 2M_{33}E_3^0 e'_{33}}{\epsilon'_{33}c_{33} + e_{33}e'_{33}}}\,\omega, \tag{22}$$

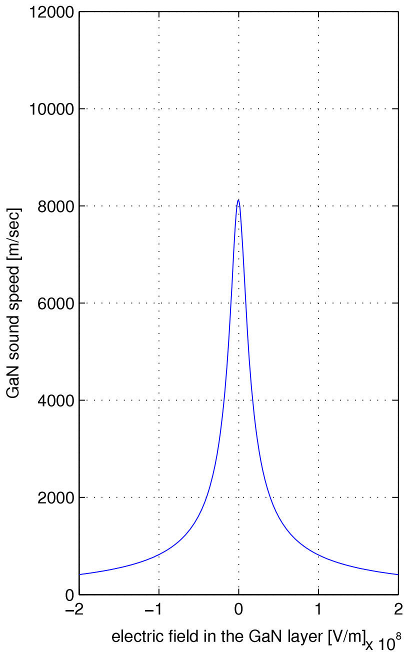

Figure 3. Plot of the effective sound speed in (left) GaN and (right) AlN as a function of the electric field.

where A and B are constant coefficients to be determined by imposing boundary conditions. Hence, the speed of sound c becomes

$$c = \frac{\omega}{k} = \sqrt{\frac{\epsilon'_{33} c_{33} + e_{33} e'_{33}}{\rho_{\mathrm{d}}(\epsilon'_{33} + 2M_{33} E_3^0 e'_{33})}}. \tag{23}$$

In figure 3 (left), the change in the effective sound speed in the GaN film layer is plotted as a function of the electric field. In actual fact, the effective sound speed decreases from 8110 m s^{-1} to approximately 46 m s^{-1} as the electric field changes from 0 to $\pm 1.5 \times 10^9$ V m^{-1}. Equation (23) reveals that this drastic change is due to the strong electrostriction value in GaN and the fact that GaN is also piezoelectric. We emphasize that such a dramatic change in sound speed and strain amplitudes with electric field will affect AC properties strongly, thus being important when considering, for example, applications of GaN quantum-well structures in semiconductor amplifiers and lasers as well as in HFET devices. Another implication is that this result may allow GaN quantum-well structures to be used in controlling sound transmission and propagation by the application of electric fields. In figure 3 (right), the electrostriction coefficient M_{zzzz} of AlN, equal to 1.46×10^{-21} [9], is used to compute the corresponding change in sound speed of wurtzite AlN versus electric field. Note that since the electrostrictive coefficient of AlN is nearly three orders of magnitude smaller than the corresponding value in GaN, the influence of electric field on sound speed is almost negligible. This result indeed shows that it is the extremely large electrostriction value of GaN which leads to the strong dependence of sound speed versus electric field. We also point out that this sound–speed effect requires the combined influence of piezoelectricity and electrostriction as evidenced by equation (23).

2.3. Resonance phenomena in a single GaN layer

Combination of equations (13) and (14) leads to an expression for the AC stress as a function of the AC strain and electric displacement:

$$\tilde{T}_3 = \left(c_{33} + \frac{e_{33}e'_{33}}{\epsilon'_{33}}\right)\tilde{S}_3 - \frac{e_{33}}{\epsilon'_{33}}\tilde{D}_3. \tag{24}$$

Let the GaN layer thickness be L and assume that stresses are zero at the GaN interfaces. Resonance conditions apply when

$$\begin{aligned} &A + B = 0, \\ &A\exp(\mathrm{i}kL) + B\exp(-\mathrm{i}kL) = 0, \end{aligned} \tag{25}$$

for $(A, B) \neq (0, 0)$, since then $\tilde{T}_3$ can be different from zero even in the absence of external currents, i.e. from equations (22) and (24) with $\tilde{D}_3 = 0$:

$$\begin{aligned} &k = \frac{n\pi}{L}, \\ &f = \frac{\omega}{2\pi} = \frac{nc}{2L} = \frac{n\sqrt{\frac{\epsilon'_{33}c_{33}+e_{33}e'_{33}}{\rho_{\mathrm{d}}(\epsilon'_{33}+2M_{33}E_3^0e'_{33})}}}{2L}, \\ &n = 1, 2, 3, \ldots. \end{aligned} \tag{26}$$

Insertion of the relevant parameters for GaN [8, 15]: $e_{33} = 0.73$ C m^{-2}, $\epsilon_{33} = 9.28\epsilon_0$, $c_{33} = 3.98 \times 10^{11}$ Pa, $\rho_{\mathrm{d}} = 6.15 \times 10^3$ kg m^{-3}, $M_{33} = 1.2 \times 10^{-18}$ m^2 V^{-2}, $L = 10$ nm, $E_3^0 = -1 \times 10^8$ V m^{-1}, and $\epsilon_0 = 8.854 \times 10^{-12}$ F m^{-1}, gives for the first resonance frequency:

$$\begin{aligned} &f_1 = 34 \text{ GHz} \quad \text{with electrostriction}, \\ &f_1 = 405 \text{ GHz} \quad \text{without electrostriction } (M_{33} = 0). \end{aligned} \tag{27}$$

Note the huge difference, due to electrostriction, even for a rather small DC electric field of 1×10^8 V m^{-1}. This result stems from the fact that the sound speed decreases drastically with increasing absolute value of the electric field as mentioned in the previous subsection. DC values of the electric field may actually approach 1×10^9 V m^{-1} in HFET applications and easily above 1×10^8 V m^{-1}, corresponding to short-circuit voltage boundary conditions [1, 4].

2.4. Resonance phenomena in a GaN/AlN heterostructure

Consider next the two-layer heterostructure depicted in figure 1 with a GaN film layer deposited on an AlN substrate. The GaN layer (layer 1) corresponds to the interval $-L_1 \leqslant z \leqslant 0$ while the AlN layer (layer 2) corresponds to the interval $0 \leqslant z \leqslant L_2$ (thus, L_1 and L_2 are positive numbers). The general solution for the strain in the two regions is (with $\tilde{D}_3 = 0$)

$$\begin{aligned} \tilde{S}_3 &= A_1\exp(\mathrm{i}k_1z) + B_1\exp(-\mathrm{i}k_1z), \\ \tilde{S}_3 &= A_2\exp(\mathrm{i}k_2z) + B_2\exp(-\mathrm{i}k_2z), \end{aligned} \tag{28}$$

with (A_i, B_i) constant coefficients and k_i the wavevectors associated with layer i. Imposing stress-free conditions at $z = -L_1$ and L_2 yields

$$\begin{aligned} B_1 &= -A_1\exp(-2\mathrm{i}k_1L_1), \\ B_2 &= -A_2\exp(2\mathrm{i}k_2L_2). \end{aligned} \tag{29}$$

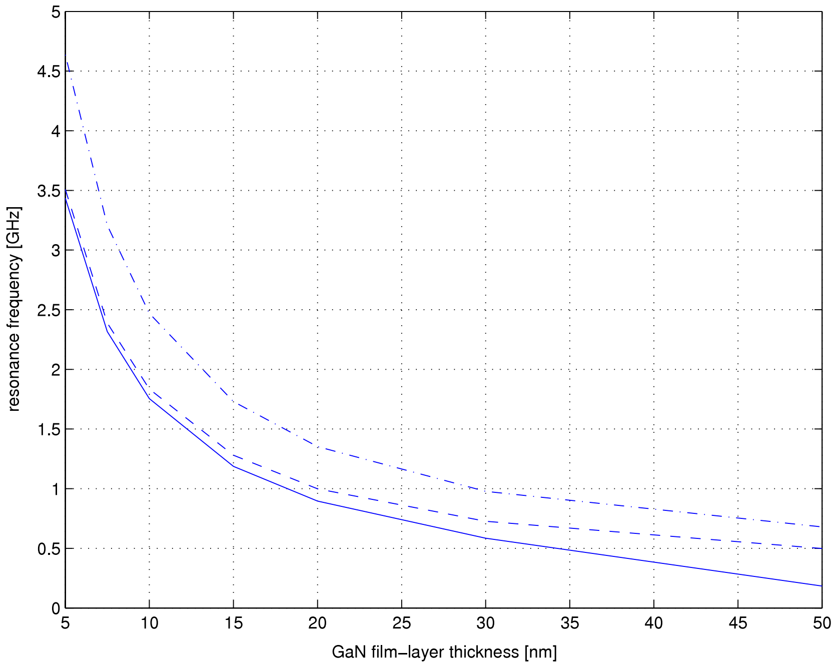

Figure 4. Plot of the first three resonance frequencies for a GaN/AlN heterostructure as a function of GaN thickness in the range 5–50 nm. The AlN layer thickness is 100 nm. The heterostructure is short-circuited.

Next, stress $\tilde{T}_3$ and the velocity $\tilde{v}_3 = \frac{1}{\mathrm{i}\omega\rho_\mathrm{d}}\frac{\partial\tilde{T}_3}{\partial z}$ are continuous everywhere in the structure and particularly at the GaN/AlN interface. Hence, from equations (24) and (29)

$$\left[c_{33}^1 + \frac{e_{33}^1 e_{33}^{1\prime}}{\epsilon_{33}^{1\prime}}\right](A_1 - A_1\exp(-2\mathrm{i}k_1L_1)) = \left[c_{33}^2 + \frac{e_{33}^2 e_{33}^{2\prime}}{\epsilon_{33}^{2\prime}}\right](A_2 - A_2\exp(2\mathrm{i}k_2L_2)),$$
$$\frac{\mathrm{i}k_1}{\rho_\mathrm{d}^1}\left[c_{33}^1 + \frac{e_{33}^1 e_{33}^{1\prime}}{\epsilon_{33}^{1\prime}}\right](A_1 + A_1\exp(-2\mathrm{i}k_1L_1)) \tag{30}$$
$$= \frac{\mathrm{i}k_2}{\rho_\mathrm{d}^2}\left[c_{33}^2 + \frac{e_{33}^2 e_{33}^{2\prime}}{\epsilon_{33}^{2\prime}}\right](A_2 + A_2\exp(2\mathrm{i}k_2L_2)),$$

where c_{33}^i is the c_{33} value in layer i, etc. The above system of two equations in A_1 and A_2 can be solved to give the resonance expressions

$$\frac{k_2}{\rho_\mathrm{d}^2}\tan(k_1L_1) = \frac{k_1}{\rho_\mathrm{d}^1}\tan(k_2L_2). \tag{31}$$

This equation can be solved so as to find the resonance frequencies for general thicknesses of the two layers in the heterostructure. Note that k_1 (GaN) and k_2 (AlN) are proportional to the frequency and determined by material constants and the DC electric field in GaN and AlN, respectively (refer to equation (22)).

In figure 4, a plot of the first three resonance frequencies is given for a GaN/AlN heterostructure as a function of the GaN film-layer thickness in the range 5–50 nm. The AlN thickness is 100 nm. Note that due to the large AlN thickness (relative to the GaN layer thickness), the first three resonance frequencies are much smaller than in the case of a single layer of GaN with thickness 10 nm. Furthermore, the resonance frequencies decrease with increasing film-layer thickness as expected.

3. Conclusions

An analysis of electrostriction in GaN-based nanostructures is presented. It is shown for a one-dimensional model that, due to large electric fields in, for example, GaN/AlN heterostructures, electrostriction plays a significant role at DC voltage conditions subject to externally imposed hydrostatic pressure or at AC conditions. In the latter case, electrostriction gives significant (sometimes even dominant) contributions to the strain distribution and resonance frequencies even in the absence of hydrostatic pressure. Finally, it must be emphasized that electrostriction calculations based on a three-dimensional model accounting for strain (piezoelectricity) are likely to reveal that electrostriction is important at DC conditions in the absence of hydrostatic pressure. The latter case is currently under consideration (work in progress) and will be published elsewhere.

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