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FLUCTUATIONS OF PIEZOELECTRIC POLARIZATION IN III-NITRIDE QUANTUM WELLS

In this work, the influence of the atomic disorder on a local piezoelectric polarization in polar II-nitride quantum wells is simulated. The calculation is performed for GaN/InGaN/GaN structures with a random distribution of In atoms in the quantum well region. The key component of the research is the valence force field model optimized for nitrides, which is used to obtain the distribution of relaxed atomic positions and the local strain tensor. The calculation showed a strong spatial non-uniformity of the piezoelectric polarization, which can even change the sign of the local polarization value and makes the distribution of a polarization potential substantially different from the standard capacitor-like picture that is observed in the case of constant polarization vectors in the quantum well and barriers.

K e y w o r d s: InGaN quantum wells, atomic disorder, piezoelectric polarization.

1. Introduction

Binary III-nitride compounds with the wurtzite-type crystal lattice have very strong piezoelectric properties. Therefore, when growing quantum structures based on InGaN/GaN heterojunctions, even a slight mismatch between the crystal lattice constants generates strong electric fields. In work [1], it was shown that those fields arise owing to both piezoelectric effects and the spontaneous polarization, which is a characteristic feature of materials with the crystal lattice of the wurtzite type.

Many physical effects associated with the polarization are known nowadays in nitride-based quantum structures. For instance, in work [2], the asymmetry of the valence bands in InN/GaN heterojunctions that were pseudomorphically grown in the (0001) direction (it is also called the *c*-direction) was associated with piezoelectric fields. It was also found that the polarization in nitrides leads to a shift of low-temperature photoluminescence spectra and an in-

crease in the lifetime of charge carriers in quantum wells [3, 4]. Some experimental works showed that the polarization in InGaN/GaN wells can be one of the physical mechanisms responsible for the non-thermal drop of a LED efficiency in the visible spectral interval [5, 6]. All those experimental facts testify to the necessity of considering the polarization effects, while theoretically simulating the properties of nitride nanostructures and devices based on them.

The theoretical works in this direction can be divided into two groups. The first group includes calculations of material constants for the spontaneous and piezoelectric polarizations, mainly from the first principles [7–9]. The second group includes studies of the influence of polarization effects on important nanostructure characteristics such as the energy band structure, overlapping of the wave functions of charge carriers, optical spectra, and charge carrier transport [10–15].

In polar InGaN/GaN quantum wells, the piezoelectric response to the tensile deformation in the *c*-direction is taken into account using the known relations [16]

$$P_z = 2\epsilon_{xx}\left(e_{31} - e_{33}\frac{C_{13}}{C_{33}}\right), \quad P_x = 0, \quad P_y = 0 \qquad (1)$$

where P_z is the component of the piezoelectric polarization vector in the *c*-direction, e_{31} and e_{33} are

components of the piezoelectric tensor, C_{13} and C_{33} are the components of the elasticity tensor, and ϵ_{xx} is the strain in the well plane. The latter parameter is usually calculated as the relative difference between the crystal lattice constants a for GaN and InGaN, $\epsilon_{xx} = (a_{\mathrm{GaN}} - a_{\mathrm{InGaN}})/a_{\mathrm{InGaN}}$.

In the vast majority of theoretical studies, the direction and magnitude of the piezoelectric polarization vector are considered to be independent of coordinates, even if local deformation fluctuations arising due to a random arrangement of atoms in the ternary InGaN compound are taken into account. A probable influence of the atomic disorder on the local piezoelectric polarization began to be studied relatively recently. In particular, in works [17, 18], substantial fluctuations of the piezoelectric polarization in III-nitride nanostructures were shown for the first time. In this work, we will continue to study the influence of the atomic disorder on a local polarization in polar InGaN/GaN wells. We consider quantum wells with various stoichiometric compositions. For the simulation of fluctuation effects, a structure with about 150,000 atoms was used. Local deformations and polarizations were calculated on the basis of the valence force field method optimized by us for nitrides with a crystal lattice of the wurtzite type. We showed that local deformations arising as a result of the atomic disorder bring about substantial non-uniformities in the piezoelectric polarization and polarization potential fields. Those non-uniformities can be responsible for experimental effects in InGaN nanostructures that cannot be explained in the framework of conventional one-dimensional models with constant polarization.

2. Theoretical Model

The key component of our research is the valence force field (VFF) model optimized for nitrides, on the basis of which the distribution of relaxed atomic positions and the local strain tensor are calculated. In its classical form, the VFF model can rather adequately describe the elastic properties in crystals with cubic symmetry. Some modifications of this model were also developed for crystals with a wurtzite-type lattice.

The variation of the interatomic bond length [19], the equilibrium geometry, and the elastic properties of binary III-nitrides and ternary compounds [20, 21] were calculated using the VFF model with two force

constants. However, all these modifications are completely unable to adequately reproduce the dynamic properties of the crystal lattice in nitrides, e.g., the phonon spectra. That is why more complicated, multiparameter approaches are applied to reproduce the static and dynamic properties of the lattice in nitrites.

In this study, we use our optimization of the VFF model for nitrides, which was described in work [22]. The potential energy of the lattice included not only the interaction energy of the nearest atoms, but also the energy of interaction with other nearest atoms, as well as a coplanar interaction. In addition, the anisotropy of the wurtzite structure was taken into account by introducing two groups of force constants: the first one describes the interatomic interaction in the c-direction, and the second one in the direction perpendicular to the c-direction. The final expression for the potential energy per atom looks like

$$
U_i = \frac{3}{16} \sum_j^{fn(i)} \alpha_{ij} (\Delta r_{ij})^2 + \frac{3}{16} \sum_m^{sn(i)} \mu_{im} (\Delta r_{im})^2 +
$$
$$
+ \frac{3}{8} \sum_j^{fn(i)} \sum_{k>j}^{fn(i)} \Bigg(\beta_{jik} (\Delta \theta r_{jik})^2 + \gamma_{jik} \Delta r_{ij} \Delta \theta r_{jik} +
$$
$$
+ \delta_{jik} \Delta r_{ij} r_{ik} + \sum_l^{\mathrm{cop}(jikl)} \nu_{ijkl} \Delta \theta r_{jik} \Delta \theta r_{ikl} \Bigg), \qquad (2)
$$

where Δr and $\Delta \theta$ are changes in the length of the interatomic bond and the angle between the bonds, respectively, due to relaxation; α, μ, β, γ, δ, and ν are force constants that determine the compressive-tensile energy of interatomic bonds with the first nearest and second nearest atoms, the bending energy of interatomic bonds, and the coplanar interaction energy. The symbols $fn(i)$, $sn(i)$, and $cop(jikl)$ denote all first and second nearest neighbor atoms to a given atom, and all atoms satisfying the coplanarity condition.

The anisotropy of the interatomic interaction leads to a set of twenty independent force constants. They were determined using a nonlinear multidimensional procedure for optimizing an objective function. The latter is the root-mean-square deviation between the calculation results for this model and known experimental data. Based on the results of this optimization, we managed to obtain a set of parameters that quite accurately reproduce both the static (the crystal lattice constants, the lengths of interatomic bonds,

the elastic constants, and the internal deformation parameters) and dynamic (the phonon spectra) properties of the crystal lattice in nitrides in the framework of a single model [22].

The model described above was applied to consider a structure consisting of two GaN barriers and an $In_xGa_{1-x}N$ well (Fig. 1). The indium atoms randomly occupied the cationic positions. The total number of atoms in the structure was rather large (about 150 000 atoms) in order not to miss fluctuation effects. The initial positions of the atoms in the structure were determined by a parallel transfer of the unit cell, whose geometry was determined by the parameters for GaN: the crystal lattice constants $a = 3.189$ Å, $c = 5.185$ Å, and the internal parameter $u = 0.3768$ [23].

Then, the procedure of the lattice potential energy minimization (relaxation) over the atomic positions and structure dimensions was carried out. As a result, the equilibrium position of each atom was obtained. The equilibrium positions of atoms were used to calculate the local strain tensor (ϵ_{ij}) for each atom [24],

$$
\begin{pmatrix} 1+\epsilon_{xx} & \epsilon_{yx} & \epsilon_{zx} \\ \epsilon_{xy} & 1+\epsilon_{yy} & \epsilon_{zy} \\ \epsilon_{xz} & \epsilon_{yz} & 1+\epsilon_{zz} \end{pmatrix} =
$$

$$
= \begin{pmatrix} R_{12,x} & R_{23,x} & R_{34,x} \\ R_{12,y} & R_{23,y} & R_{34,y} \\ R_{12,z} & R_{23,z} & R_{34,z} \end{pmatrix} \begin{pmatrix} R^0_{12,x} & R^0_{23,x} & R^0_{34,x} \\ R^0_{12,y} & R^0_{23,y} & R^0_{34,y} \\ R^0_{12,z} & R^0_{23,z} & R^0_{34,z} \end{pmatrix}^{-1}, \quad (3)
$$

where $\mathbf{R}^0_{12}$, $\mathbf{R}^0_{23}$, $\mathbf{R}^0_{34}$, and $\mathbf{R}_{12}$, $\mathbf{R}_{23}$, $\mathbf{R}_{34}$ are vectors defining – before and after the relaxation procedure, respectively – the geometry of a tetrahedron, the vertices of which are occupied by nearest neighbor atoms (Fig. 1).

Afterward, the piezoelectric polarization vector field was calculated from the linear relationship between the polarization and deformation components,

$$
\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & e_{15} & 0 \\ 0 & 0 & 0 & e_{15} & 0 & 0 \\ e_{31} & e_{31} & e_{34} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \epsilon_1 \\ \epsilon_2 \\ \epsilon_3 \\ \epsilon_4 \\ \epsilon_5 \\ \epsilon_6 \end{pmatrix}, \quad (4)
$$

where the strain tensor components are written in the matrix notation ($\epsilon_1 = \epsilon_{xx}$, $\epsilon_2 = \epsilon_{yy}$, $\epsilon_3 = \epsilon_{zz}$, $\epsilon_4 = 2\epsilon_{yz}$, $\epsilon_5 = 2\epsilon_{xz}$, $\epsilon_6 = 2\epsilon_{xy}$). The piezoelectric constants for $In_xGa_{1-x}N$ were found by linearly interpolating between the values for GaN ($e_{33} = 0.75$ C/m^2,
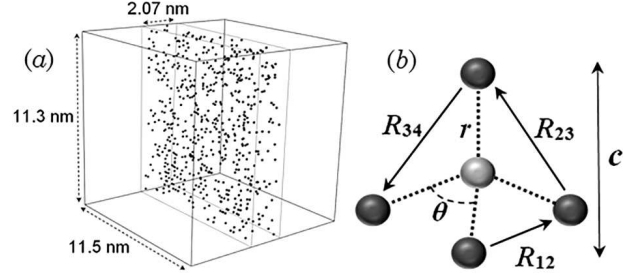

Fig. 1. Schematic diagram of a 2.07-nm-thick quantum well for which the calculations were performed (points demonstrate one of the random distributions of indium atoms in the well) (*a*). The arrangement of the nearest neighbor atoms in the hexagonal crystal structure and the vectors defining the geometry of this arrangement (*b*)

$e_{31} = -0.44$ C/m^2, and $e_{15} = -0.31$ C/m^2) and InN ($e_{33} = 1.14$ C/m^2, $e_{31} = -0.59$ C/m^2, and $e_{15} = -0.43$ C/m^2) [9].

Since the polarization affects the physical properties of systems through the electric field, at the final stage of our simulation, we found the distribution of the polarization potential. For this purpose, the point dipole method was used. Each cationic atom, when shifting with respect to its four nearest neighbor anionic atoms due to a deformation, violates the charge symmetry and creates an electric field similar to that of a dipole. In other words, each i-th cation atom – or, more precisely, each atomic tetrahedron covering this cation – was assigned a dipole moment, which was defined as $\mathbf{d}_i = \mathbf{P}_i \cdot V_i$, where V_i is the atomic tetrahedron volume, and $\mathbf{P}_i$ is the local polarization vector at the cation atom's location. The resulting potential field was found as the sum of all cation dipole fields,

$$
\phi(\mathbf{r}) = \frac{1}{4\pi\epsilon\epsilon_0} \sum_i \frac{\mathbf{d}_i\,(\mathbf{r} - \mathbf{r}_i)}{|\mathbf{r} - \mathbf{r}_i|^3}, \quad (5)
$$

where ϵ is the dielectric constant of the material ($\epsilon \approx 9.5$ for GaN [25])

3. Discussion of Results

The calculations were carried out for a GaN/$In_xGa_{1-x}N$/GaN structure $11.5 \times 11.3 \times 6.2$ nm^3 in dimensions with a 2.07-nm-thick quantum well (4 crystal lattice constants in the c-direction). In the framework of our research, the most difficult and time-consuming stage was the minimization procedure for the potential energy of the structure.
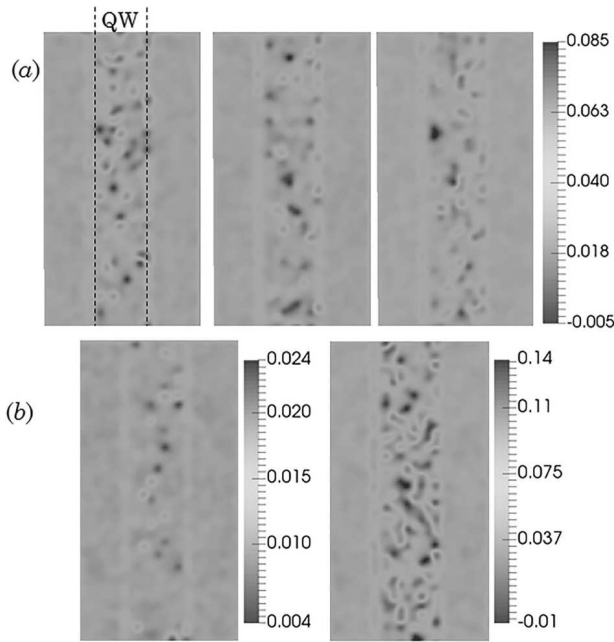
Fig. 2. Spatial distributions of the P_z-projection of the piezo-electric polarization in $In_xGa_{1-x}N$ quantum wells along the planes parallel to the c-direction (the dashed lines in the first figure show the quantum well boundaries): $x = 0.3$ (distributions in three various planes) (*a*), $x = 0.1$ (*b*), and $x = 0.5$ (*c*). The right-hand scales for the P_z-values are given in C/m^2 units
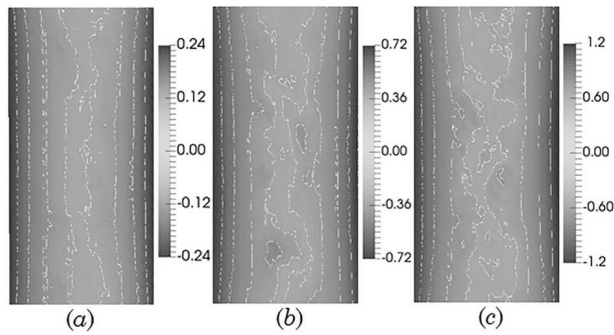

Fig. 3. Spatial distributions of the polarization potential over the plane parallel to the c-direction in $In_xGa_{1-x}N$ quantum wells for $x = 0.1$ (*a*), 0.3 (*b*), and 0.5 (*c*). White lines are used to show the shape of equipotential surfaces. The right-hand scales for the potential values are given in Volt units

The numerical determination of the global minimum of the potential energy as a function of the atomic positions and the structure size became complicated owing to the presence of plenty of subsidiary local minima. Furthermore, because of the huge number of degrees of freedom, the classical conjugate gra-

dient algorithm of minimization requires too much time, already if the number of atoms exceeds a value of about 50 000. Therefore, for large structures, we used an approach based on the polytope algorithm [26]. This algorithm only needs the calculation of the potential energy itself rather than its gradient, which speeds up the procedure of finding just the global minimum for highly nonlinear functionals. After the minimization, we calculated the patterns of deformation fields and the piezoelectric polarization.

Note that, in order to be in agreement with experimental conditions at the pseudomorphic growing of InGaN quantum wells in the c-direction, a strain resulting from the difference between the crystal lattice constants in the substrate and the quantum structure itself was also taken into account when calculating the strain fields. This strain can be called external in the sense that it has no relation to the local effects of the atomic disorder in the well. Specifically, the external strain was introduced into our model by inserting a component (matrix), which is constant and independent of the atomic positions, into Eq. (3). Thus, the piezoelectric polarization fields were a result of both external strains and local strains arising due to the atomic disorder.

In this work, we do not consider the spontaneous polarization. The magnitude of the spontaneous polarization in quantum wells is determined by the difference between the spontaneous polarization constants of the materials of the well itself and the barriers. Since the polarization constants in binary GaN and InN are very close, this fact makes it possible to assume the spontaneous polarization in InGaN/GaN quantum wells to be much weaker than the piezoelectric one [27].

Figure 2, *a* illustrates the polarization patterns – or, more precisely, the projections of the polarization vector along the c-direction – in $In_xGa_{1-x}N$/GaN quantum wells with $x = 0.3$ in some different planes parallel to the c-direction. As we see from the figure, the usual picture of a uniform piezoelectric polarization field, which is assumed by formula (1), is not observed in this case. Instead, substantial polarization fluctuations are clearly visible, with the size of their spatial localization regions not exceeding one crystal constant (i.e., less than 5 Å), as a rule. Such a non-uniformity of the polarization field can be associated with local deformations induced by random arrangements of indium atoms in the quantum well region.

The determining role of the atomic disorder is confirmed, in particular, if we compare the polarization distributions in the quantum well with various concentrations of indium atoms (Figs. 2, *b* and 2, *c*). The higher the concentration of indium atoms, the larger the fluctuations, up to the change of the polarization sign (direction). The polarization non-uniformity immediately along the interface between the barriers and the well can be a result of the transverse deformations arising due to the mismatch between the lattice constants in the barrier and well materials.

In mathematical models, the influence of a polarization on the electronic states of charge carriers in quantum structures can be taken into account by summing up the polarization potential, the periodic potential of the crystal lattice, and the quantum confinement potential in the Schrödinger equation. It is obvious that the revealed polarization fluctuations will be reflected on the polarization potential field as well. Our calculations on the basis of the point dipole model gave the results depicted in Fig. 3. Note that Fig. 3 illustrates the potential distribution arising due to the piezoelectric polarization only, without account for the potential induced by the space charge region.

General conclusions that can be drawn from those results are as follows. Even at low concentrations of indium atoms in the quantum well, the equipotential surfaces are not planes parallel to the *c*-direction, as it is assumed in the vast majority of mathematical models for InGaN/GaN nanostructures. One can also see from Fig. 3 that the higher the concentration of indium atoms, the more distinct the polarization potential distribution from the ordinary capacitor-like picture obtained for constant polarization vectors in the well and barriers.

4. Conclusions

The obtained results show that the atomic disorder in quantum wells based on the InGaN/GaN polar structures gives rise to the appearance of local deformations. Such deformations induce substantial fluctuations of the piezoelectric polarization field. The calculations have shown that, in the case of quantum wells with a high concentration of indium atoms, the local polarization can even change its direction. Polarization fluctuations correspondingly affect the polarization potential distribution. The one-dimensional approximation, which is often used for modeling the physical properties of InGaN/GaN quantum struc-

tures under polarization conditions, does not make allowance for any fluctuation effects. That is why the development of more complicated three-dimensional models to analyze the influence of polarization fluctuations on the recombination and transport properties of charge carriers in quantum wells and nanostructures in general is of great interest.

In this work, possible fluctuations of the spontaneous polarization were not taken into account. However, the inclusion of the spontaneous local polarization in bulk compounds or one-dimensional quantum structures based on III-nitrides into the proposed model is of considerable interest and may be the subject of a further research.

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ФЛУКТУАЦІЇ П'ЄЗОЕЛЕКТРИЧНОЇ
ПОЛЯРИЗАЦІЇ В КВАНТОВИХ ЯМАХ
НА ОСНОВІ III-НІТРИДІВ

В роботі змодельовано вплив атомної невпорядкованості на локальну п'єзоелектричну поляризацію в полярних квантових ямах на основі III-нітридів. Розрахунок проводився для структур GaN/InGaN/GaN з випадковим розподілом атомів In в області квантової ями. Ключовою складовою нашого дослідження є оптимізована для нітридів модель валентних силових полів, на основі якої ми отримували розподіл релаксованих атомних положень та локальний тензор напруг. Розрахунок показав сильну просторову неоднорідність п'єзоелектричної поляризації, при якій локальне значення поляризації може навіть змінювати свій знак, а розподіл поляризаційного потенціалу значно відрізняється від традиційної картини конденсаторного поля в умовах постійних векторів поляризації в ямі та бар'єрах.

К л ю ч о в і   с л о в а: InGaN квантові ями, атомна невпорядкованість, п'єзоелектрична поляризація.