Theory of second-order piezoelectric fields in III-N nanostructures

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***Abstract*—We present a detailed analysis of second-order piezoelectric fields in wurtzite nitride nanostructures. A symmetry adapted approach is used to describe these fields as a function of the growth plane. The derived analytic expression can easily be implemented in existing codes for quantum dots and wells. In terms of the field strength, our results reveal that when including second-order effects an improved agreement between theory and experiment is achieved. Our calculations indicate that in nonpolar InGaN/GaN dots, second-order effects can further reduce residual first-order related fields. In c-plane systems, second-order effects become significant with increasing In content.**

***Index Terms*—nitrides, second-order piezoelectric fields**

I. Introduction

Wurtzite nitride semiconductors GaN, AlN, InN and their respective alloys have attracted considerable interested for optoelectronic devices [1]. However, when realizing c-plane heterostructures such as quantum dot (QDs) and wells (QWs), these systems exhibit intrinsic electrostatic built-in fields of the order of MV/cm [1], which lead to a strong reduction of the radiative recombination rate due to a spatial separation of carrier wave functions. To decrease or ideally eliminate these fields, several techniques have been discussed in the literature, e.g. growth on non-c-planes (semi- and nonpolar) [1]. Thus, a detailed understanding of how the built-in field changes with growth plane is required. So far, most theoretical studies include spontaneous and *first-order* strain related piezoelectric fields only. Here, we address *second-order* piezoelectric effects in wurtzite nitride-based QWs and QDs [4], [3]. We present analytic expressions for first- and second-order piezoelectric polarization vector fields as a function of the incline angle θ to the c-axis [3]. These expressions are easy to implement in existing codes, thus allowing to study the impact of second-order piezoelectricity on built-in fields in c- and non-c-plane QWs and QDs. Our results reveal an improved agreement between experimental and theoretical built-in field values in QWs when including second-order effects in the calculations. Also, we find that with increasing In content, second-order effects become significant in InGaN QDs and modify the electronic and optical properties of these systems noticeably [4]. We show that this becomes important when targeting red emitting InGaN QDs.

II. Theoretical Framework

To describe piezoelectric polarization vector fields in wurtzite semiconductors as a function of the growth plane, we proceed in the following way. First, the expressions for first- and second-order polarization vector fields need to be known in a reference coordinate system. We use here the "standard" coordinate system where the c-axis is parallel to the z-axis of the coordinate system; equations for spontaneous and first-order piezoelectric polarization are well established. This is not the case for second-order piezoelectricity. We build on Ref. [2], where the non-vanishing and identical coefficients have been deduced. Our derived expressions are given in Ref. [3]. Having established these equations, using an rotation matrix U, we transform the expressions given in the coordinate system (x, y, z) (z-axis parallel to c-axis) to a primed coordinate system (x', y', z'), which is then characterized by the incline angle θ to the c-axis. In our case the matrix U describes a rotation around the y-axis. Therefore, the z-axis of the simulation cell is always oriented along the growth direction of the system under consideration. The derived analytic expressions can easily be implemented in existing QW and QD codes. Thus, a symmetry adapted description of the built-in field in wurtzite nanostructures is achieved. Once the polarization vector fields are known as a function of θ, Poisson's equation is solved to obtain the corresponding built-in potential in a heterostructure. More details are given in Ref. [3].

III. Results

A. Quantum wells

To study the impact of second-order piezoelectric effects on the built-in field in InGaN/GaN QWs, we have compared our calculated field values with experimental data (F_{exp}). In our calculations we have chosen the same structural parameters (well width, barrier thickness, In content) as the experiment and performed calculations in the absence ($F^{\text{SP+FO}}$) and in the presence of second-order effects ($F^{\text{SP+FO+SO}}$). The results are summarized in Table I. Overall, we find that with including second-order piezoelectric effects the agreement between theory and experiment is improved when compared to a first-order only calculation. Similar results are found for GaN/AlGaN systems [3]. In general, when comparing the calculated fields to the experimental data, we observe that with first-order piezoelectricity only the theory underestimate the magnitude of the electric field in the respective structures.

Our theoretical studies also show that especially for semipolar QWs with high incline angle values ($55^\circ \leq \theta \leq 80^\circ$ and

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TABLE I
COMPARISON BETWEEN LITERATURE EXPERIMENTAL DATA (F_{EXP}) AND OUR CALCULATED (F_{THEO}) ELECTRIC FIELD VALUES IN INGAN QWS. THEORETICAL RESULTS INCLUDING FIRST-ODER PIEZOELECTRIC AND SPONTANEOUS POLARIZATION ONLY ARE DENOTED BY $F_{\mathrm{THEO}}^{\mathrm{SP+FO}}$; RESULTS WITH SECOND-ORDER EFFECTS INCLUDED ARE GIVEN BY $F_{\mathrm{THEO}}^{\mathrm{SP+FO+SO}}$.

$In_xGa_{1-x}N$/GaN MQWs	θ (degree)	F_{exp} (kV/cm)	$F_{\mathrm{theo}}^{\mathrm{SP+FO}}$ (kV/cm)	$F_{\mathrm{theo}}^{\mathrm{SP+FO+SO}}$ (kV/cm)
x=0.12	0	1600 [5]	1530	1620
x=0.12	58	-575±150 [6]	-397	-444
x=0.15	118	840±150 [6]	587	666

$105° \leq \theta \leq 120°$), second-order piezoelectricity noticeably contributes to the overall built-in field [3]. For example, in a semipolar $(11\bar{2}2)$ $In_{0.22}Ga_{0.78}N$/GaN QW system ($\theta \approx 58°$), second-order effects increase the magnitude of the electric field by approximately 20% when compared to a calculation where second-order piezoelectricity is neglected.

B. Quantum dots

In addition to QWs, we have also performed calculations for c-plane QDs [4]. To investigate the impact of second-order effects on the electronic and optical properties of these system, we have combined our symmetry adapted polarization field model with a symmetry adapted $\mathbf{k} \cdot \mathbf{p}$ framework, implemented in the software package S/Phi/nX. Using this approach we have evaluated the emission wavelength λ of a lens-shaped InGaN/GaN QD as a function of the In content. These calculations have been carried out in the absence, only spontaneous and first-order piezoelectric polarization ($\lambda^{\mathrm{FO+SP}}$) included, and in the presence of second-order effects (λ^{Tot}). The calculated emission wavelength shift, $\Delta\lambda = \lambda^{\mathrm{Tot}} - \lambda^{\mathrm{FO+SP}}$, is displayed in Fig. 1. As one can see, for In contents up to 20%, emission wavelength shifts due to second-order effects are small ($\Delta\lambda \leq 10$ nm). But, when the In content exceeds 30%, significant changes in $\Delta\lambda$ are observed. For instance, for an InGaN QD with 40% In, second-order effects induce a wavelength shift of approximately 50 nm. Overall, our study shows that second-order piezoelectricity leads to emission at longer wavelength when compared to a "standard" first-order calculation. This finding is important since experimentally red-emitting InGaN/GaN QDs have recently been targeted and In contents of around 40% have been realized [7]. Thus when aiming for emission in this spectral range, second order effects should be taken into account for an accurate description of the system. Furthermore, due to the second-order induced wavelength shift, lower In contents should be required to achieve red emission. While this indicates a positive effect on the interrelation of In content and emission wavelength, we find that the radiative lifetime is increased by second-order effects, which is detrimental for efficient light emission [4].

We have also analyzed the impact of second-order piezoelectricity on the built-in field in non-c-plane nitride (GaN and InGaN) systems[3]. Our calculations indicate that for GaN/AlGaN structures second-order effects are of secondary importance; here the built-in potential is dominated by spontaneous polarization. However, for InGaN-based QDs we find

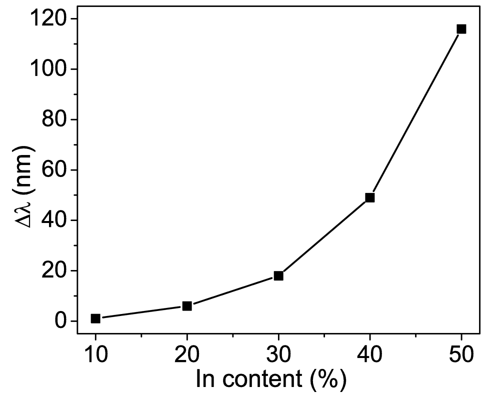

Fig. 1. Wavelength shift $\Delta\lambda$ due to second-order piezoelectric effects in a lens-shaped $In_xGa_{1-x}N$/GaN QD (base diameter d=14 nm; height h=3 nm) as a function of the In content. More details are given in the text.

that for instance in a non-polar $In_{0.25}Ga_{0.75}N$/GaN dot second-order effects can cancel first-order related contributions, leading to an almost field free situation.

IV. CONCLUSION

We have presented a symmetry adapted approach to describe second-order piezoelectric effects in c- and non-c-plane nitride heterostructures. The derived analytic expressions can easily be implemented in existing built-in field codes. Our results show that when including second-order effects in the model an improved agreement between theory and experiment is achieved in terms of the magnitude of the built-in field. Also, we find that second-order piezoelectricity becomes increasingly important in InGaN/GaN QDs with increasing In content and can lead to significant emission wavelength shifts as well as an increase in the radiative lifetime.

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