Significance of third-order elasticity for determination of the pressure coefficient of the light emission in strained quantum wells

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We investigate the contribution arising from third-order elasticity to the pressure coefficient of the light emission (dE_E/dP) in strained zinc-blende InGaAs/GaAs and InGaN/GaN quantum wells (QWs) grown in a (001) direction. In the framework of the third-order elasticity theory, we develop a model of pressure tuning of strains in these structures, which is then used to determine the coefficient dE_E/dP. In the calculations of dE_E/dP, we use a consistent set of the second- and third-order elastic constants which has been obtained from *ab initio* calculations. Our results indicate that the usage of third-order elasticity leads to significant reduction in dE_E/dP in strained (001)-oriented InGaAs/GaAs and InGaN/GaN QWs, in comparison to the values of dE_E/dP obtained by using the linear theory of elasticity. In the case of InGaAs/GaAs QWs, the values of dE_E/dP calculated using third-order elasticity are in reasonable agreement with experimental data. For InGaN/GaN QWs, better agreement between theoretical and experimental values of dE_E/dP is obtained when instead of third-order elasticity, pressure dependence of the second-order elastic constants is taken into account.

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In recent years, semiconductor quantum structures, including quantum wells (QWs) and quantum dots (QDs), have been extensively studied under hydrostatic pressure.[1–5] These studies have revealed that pressure coefficient of the light emission (dE_E/dP) for a quantum structure can be substantially different from the pressure coefficient of the band gap for unstrained bulk material of the same chemical composition. It has been found that the magnitude of dE_E/dP in quantum structures depends significantly on the internal misfit strains and the built-in electric fields.[1–3] The linear theory of elasticity and piezoelectricity often failed to predict the values of dE_E/dP in quantum structures, and the nonlinear elastic and piezoelectric effects have been invoked.[2–6]

Anomalously small values of dE_E/dP were initially reported for compressively strained (001)-oriented InGaAs/GaAs QWs.[1] Wilkinson *et al.*[1] found that in these structures, dE_E/dP decreases rapidly with increasing the In concentration in the QWs. They claimed that neither the linear theory of elasticity nor the third-order elasticity theory was able to reproduce the experimental results. Afterward, Sly and Dunstan[7] found significant dependence of dE_E/dP on the misfit strain in InGaAs/GaAs QWs, which could not be accounted for by the linear theory of elasticity. Later on, Frogley *et al.*[2] proposed to describe these findings using the nonlinear elasticity theory, in which pressure dependence of elastic constants was taken into account. Since that time, the pressure dependence of elastic constants has been used in determination of dE_E/dP in various quantum structures, including (111)-oriented InGaAs/GaAs QWs,[8] InAs/GaAs QDs,[4] (001)-oriented zinc-blende InGaN/GaN QWs,[9] and (0001)-oriented wurtzite InGaN/GaN and GaN/AlGaN QWs.[10]

The purpose of this work is to examine in depth the contribution arising from third-order elasticity to dE_E/dP in strained (001)-oriented InGaAs/GaAs and InGaN/GaN QWs. In these structures, the built-in electric field is absent and the influence of third-order elasticity on dE_E/dP can be studied without any interaction with the piezoelectric effect. Obviously, we assume that the QWs are coherently grown on the substrates or the buffer layers, with no interfacial disorder and compositional and well-width fluctuations such that the continuum theory of elasticity can be applied. In order to reach the goal, we first derive expressions for pressure tuning of strains in a (001)-oriented zinc-blende QW using the third-order elasticity theory. Then, we perform *ab initio* calculations of the second- and third-order elastic constants for InAs and GaAs. (For zinc-blende InN and GaN, we use the elastic constants reported in Ref. 11.) Finally, we calculate dE_E/dP for strained (001)-oriented InGaAs/GaAs and InGaN/GaN QWs. The obtained results indicate that the usage of third-order elasticity causes significant reduction in dE_E/dP in the considered structures and improves considerably the agreement with experimental data.

Let us start from the definition of the elastic energy density of a crystal under external hydrostatic pressure P, with terms up to the third order in strains,

$$U = \frac{1}{2}\sum_{i,j=1}^{6} C_{ij}\eta_i\eta_j + \frac{1}{6}\sum_{i,j,k=1}^{6} C_{ijk}\eta_i\eta_j\eta_k + PdV, \qquad (1)$$

where C_{ij} and C_{ijk} are the second- and third-order elastic constants, respectively; η_α is the tensor of Lagrangian strain in the Voigt convention ($\eta_1 \rightarrow \eta_{xx}$, $\eta_2 \rightarrow \eta_{yy}$, $\eta_3 \rightarrow \eta_{zz}$, $\eta_4 \rightarrow 2\eta_{yz}$, $\eta_5 \rightarrow 2\eta_{xz}$, $\eta_6 \rightarrow 2\eta_{xy}$).[12] For a (001)-strained zinc-blende crystal, the off-diagonal elements of $\eta_{\alpha\beta}$ vanish and U takes the following form:

$$\begin{aligned} U = {} & \frac{1}{2}C_{11}(\eta_{xx}^2 + \eta_{yy}^2 + \eta_{zz}^2) + C_{12}(\eta_{xx}\eta_{yy} + \eta_{yy}\eta_{zz} + \eta_{xx}\eta_{zz}) \\ & + C_{123}\eta_{xx}\eta_{yy}\eta_{zz} + \frac{1}{6}C_{111}(\eta_{xx}^3 + \eta_{yy}^3 + \eta_{zz}^3) + \frac{1}{2}C_{112}(\eta_{xx}^2\eta_{yy} \\ & + \eta_{xx}^2\eta_{zz} + \eta_{xx}\eta_{yy}^2 + \eta_{xx}\eta_{zz}^2 + \eta_{yy}\eta_{zz}^2 + \eta_{yy}^2\eta_{zz}) \\ & + P(\sqrt{(1+2\eta_{xx})(1+2\eta_{yy})(1+2\eta_{zz})} - 1). \end{aligned} \qquad (2)$$

The diagonal components of Lagrangian stress are derivatives of U with respect to $\eta_{\alpha\alpha}$, which gives

TABLE I. Second- and third-order elastic constants (in GPa) for zinc-blende GaAs and InAs.

	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{155}	C_{456}
GaAs	100	50	52	−587	−346	−74	−19	−248	−18
	(100)[a]	(49)[a]	(52)[a]	(−561)[a]	(−337)[a]	(−83)[a]	(−14)[a]	(−244)[a]	(−22)[a]
InAs	72	43	33	−404	−268	−121	−5	−138	−6

[a]Reference 11.

$$t_{zz} = C_{11}\eta_{zz} + C_{12}(\eta_{xx} + \eta_{yy}) + \frac{1}{2}C_{111}\eta_{zz}^2 + C_{112}\left[\eta_{zz}(\eta_{xx} + \eta_{yy}) + \frac{1}{2}(\eta_{xx}^2 + \eta_{yy}^2)\right] + C_{123}\eta_{xx}\eta_{yy} + P\sqrt{(1+2\eta_{xx})(1+2\eta_{yy})/(1+2\eta_{zz})}. \quad (3)$$

[From Eq. (3), one can obtain t_{xx} and t_{yy} by applying cyclic permutations $x \to y \to z$.] Now, using the constrained conditions for a (001)-oriented QW, $\eta_{xx} = \eta_{yy} \equiv \eta_{\|}$ and $t_{zz} = 0$,[13] we find

$$0 = C_{11}\eta_{zz} + 2C_{12}\eta_{\|} + \frac{1}{2}C_{111}\eta_{zz}^2 + C_{112}(2\eta_{\|}\eta_{zz} + \eta_{\|}^2) + C_{123}\eta_{\|}^2 + P(1+2\eta_{\|})/\sqrt{1+2\eta_{zz}}. \quad (4)$$

Solving Eq. (4) numerically, one obtains η_{zz} as a function of $\eta_{\|}$. Since the off-diagonal elements of $\eta_{\alpha\beta}$ vanish, the in-plane Lagrangian strain $\eta_{\|}$ can be expressed by the in-plane usual strain $\varepsilon_{\|}$ (i.e., the in-plane strain, which is defined in the infinitesimal theory of elasticity) using

$$\eta_{\|} = \varepsilon_{\|} + \frac{1}{2}(\varepsilon_{\|})^2 \quad (5)$$

and

$$\varepsilon_{\|} = \frac{a_{\mathrm{sub}}(P)}{a_{\mathrm{QW}}} - 1, \quad (6)$$

where a_{QW} is the bulk lattice constant of unstrained QW material and $a_{\mathrm{sub}}(P)$ is the pressure-dependent lattice constant of the substrate.[14] For InGaAs/GaAs QWs, the thickness of the substrate is much larger the QW width; thus, one can determine $a_{\mathrm{sub}}(P)$ by

$$a_{\mathrm{sub}}(P) = a_{\mathrm{sub},0}\left(1 - \frac{P}{C_{11,\mathrm{sub}} + 2C_{12,\mathrm{sub}}}\right), \quad (7)$$

where $a_{\mathrm{sub},0}$ and $C_{11,\mathrm{sub}}$ and $C_{12,\mathrm{sub}}$ are the lattice constant and the elastic constants of an unstrained GaAs substrate, respectively. In the case of InGaN/GaN QWs, determination of pressure tuning of $\varepsilon_{\|}$ is much more complicated since zinc-blende GaN substrates are unavailable. The zinc-blende InGaN/GaN QWs are usually grown on GaN buffer layers which are attached to SiC substrates through nonpseudomorphic layers with high density of dislocations and of unknown elastic properties.[9,15] In order to describe the pressure tuning of strains in such complicated structures, the pressure-dependent effective lattice constant is commonly introduced by

$$\tilde{a}_{\mathrm{sub}}(P) = a_{\mathrm{buf},0}\left(1 - \frac{P}{3B_{\mathrm{sub}}}\right), \quad (8)$$

where $a_{\mathrm{buf},0}$ is the lattice constant of an unstrained GaN and B_{sub} is the bulk modulus of SiC substrate.[9,10,16]

Using Eqs. (5)–(8), we determine $\eta_{\|}$ as a function of P and then, solving numerically Eq. (4), we obtain the pressure dependence of η_{zz}. The last step is to determine the pressure dependence of the zz component of usual strain tensor ε_{zz}. For (001)-oriented QWs, the following formula applies:

$$\varepsilon_{zz} = \sqrt{1+2\eta_{zz}} - 1. \quad (9)$$

Next, we link pressure-dependent strains $\varepsilon_{\|}$ and ε_{zz} to effective-mass Hamiltonians for conduction and valence bands and solve the effective-mass Schrödinger equations for certain values of P. In this way, we determine the pressure dependence of the fundamental optical transition energy $E_E(P)$, which allows us to calculate the value of the coefficient dE_E/dP.

Now, let us specify the values of material parameters which will be used in calculations of dE_E/dP in InGaAs/GaAs and InGaN/GaN QWs. We focus on determination of the second- and third-order elastic constants. Recently, it has turned out that calculations based on the density-functional theory (DFT) with generalized gradient approximation (GGA) for the exchange-correlation functional provide reliable predictions of the C_{ij} and C_{ijk} elastic constants in semiconductors.[11] Using the GGA-DFT approach, the theoretical values of C_{ij} and C_{ijk} have been determined for Si, GaAs, and zinc-blende nitrides.[11] Reasonable agreement with experimental results has been obtained for the elastic constants of Si and GaAs, although the GGA-DFT calculations suffer from the well-known shortcoming connected with overestimation of the equilibrium lattice constant of solids.[11] Unfortunately, the elastic constants for InAs were not calculated in Ref. 11. Therefore, we have performed calculations of C_{ij} and C_{ijk} for InAs and GaAs. Our approach is based on the DFT calculations of the total energy of a crystal as a function of Lagrangian strain. The total-energy calculations have been performed using the VASP code[17] for six types of distortions listed in Ref. 11. Similarly to Ref. 11, the GGA approximation for the exchange-correlation functional has been applied.[18] We have chosen the kinetic-energy cutoff to be equal to 800 eV, and the Brillouin-zone integrations have been computed using a $15 \times 15 \times 15$ MONKHORST-PACK k-space grid. The results are presented in Table I. One can see that our values of the elastic constants for GaAs are in very good agreement with the results found in Ref. 11. Therefore, we accept the obtained predictions of C_{ij} and C_{ijk}

for InAs. For InGaAs and InGaN alloys, we assume a linear dependence of the C_{ij} and C_{ijk} elastic constants on the In concentration. The *ab initio* study of the composition dependence of C_{ij} and C_{ijk} in the ternary alloys is beyond the scope of this work.[19] The other material parameters which are used in calculations of dE_E/dP have been taken from Refs. 2 and 20–23. Note that for InGaN alloys, we use a nonlinear dependence of the hydrostatic deformation potential a_H on the composition which has been determined from *ab initio* calculations in Ref. 22. Unfortunately, no reliable data on a_H in InGaAs alloys appear to exist.[20] Therefore, we use linear interpolation of a_H in this case. The energy gaps of the QWs have been calculated assuming a bowing parameter of −0.477 for InGaAs and −1.37 for InGaN, respectively.[20,23]

Finally, we present and discuss the results of calculations of dE_E/dP in strained (001)-oriented InGaAs/GaAs and InGaN/GaN QWs. In order to investigate the contribution to dE_E/dP originating from third-order elasticity, we compare the values of dE_E/dP which have been calculated using third-order elasticity with the results obtained using the linear (second-order) theory of elasticity and with experimental data taken from the literature. In the case when the linear theory of elasticity has been applied, we have determined the pressure dependence of ε_{zz} using the well-known formula (see Refs. 9 and 10)

$$\varepsilon_{zz} = -2\frac{C_{12}}{C_{11}}\varepsilon_{\parallel} - \frac{P}{C_{11}}. \tag{10}$$

Considering InGaAs/GaAs system, we have performed calculations of dE_E/dP for the QWs which were studied experimentally in Ref. 1. Note that the experimental values of dE_E/dP were determined by measuring pressure dependences of photoluminescence peak energies for the QWs relative to the bulk GaAs substrate.[1] In this way, the possible error in pressure calibration and unwanted influence of nonlinear dependence of $E_E(P)$ on dE_E/dP were minimized.[1,2] Regarding the theoretical values of dE_E/dP, we have determined them from computed dependences of $E_E(P)$ using the finite difference formula

$$dE_E/dP \cong \frac{E_E(\Delta P) - E_E(0)}{\Delta P}. \tag{11}$$

Relatively small value of $\Delta P = 0.25$ GPa has been chosen in order to reach the consistent determination of dE_E/dP with the experimental procedure used in Ref. 1. In Fig. 1, we demonstrate the values of dE_E/dP which have been obtained for 10 nm InGaAs/GaAs QWs as functions of In concentration. (The experimental results from Ref. 1 have been rescaled according to suggestions found in Ref. 2.) Not surprisingly (see Refs. 1 and 2), the values of dE_E/dP obtained using the linear theory of elasticity are larger than the experimental data. The usage of third-order elasticity improves determination of the pressure tuning of strains in the QWs which leads to significant decrease in dE_E/dP. Importantly, the contribution to dE_E/dP arising from third-order elasticity increases with increasing the In concentration in the QWs, which considerably enhances the agreement between the theoretical and experimental results.

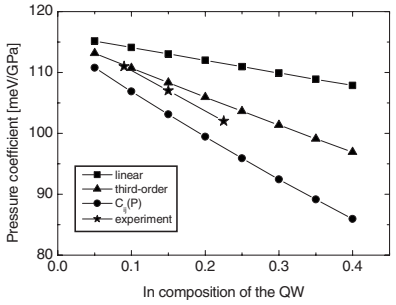

FIG. 1. Values of dE_E/dP for 10 nm InGaAs/GaAs QWs as functions of In concentration. Stars represent the experimental results (see Refs. 1 and 2). Squares, triangles, and circles correspond to the theoretical results which are obtained using (i) the linear theory of elasticity, (ii) the third-order elasticity theory, and (iii) nonlinear elasticity originating from the pressure dependence of the second-order elastic constants, respectively. Solid lines are added to guide the eyes.

In Fig. 1, we have also presented the values of dE_E/dP calculated assuming the dependence of the second-order elastic constants on hydrostatic pressure. In this case, we have calculated pressure tuning of strains in the QW using Eqs. (6), (7), and (10) and assuming that the elastic constants of GaAs and InAs change with the total built-in hydrostatic pressure (P_{tot}) by

$$C_{ij}(P_{\text{tot}}) = C_{ij} + C'_{ij}P_{\text{tot}}, \tag{12}$$

where C'_{ij} are the pressure derivatives of the elastic constants and indices take the values $(ij)=(11),(12),(44)$, respectively. The values of C'_{ij} are determined from the elastic constants C_{ij} and C_{ijk} using the formulas presented in Ref. 11. Note that the values of P_{tot} are different in the substrate than in the QW since the QW is compressively strained even at ambient pressure.[10] In the substrate, P_{tot} takes the same values as external hydrostatic pressure P. For the QW, P_{tot} is determined by solving the following transcendental equation (see Ref. 10):

$$P_{\text{tot}} = P - \frac{2}{3}\left\{\varepsilon_{\parallel}\left[C_{11}(P_{\text{tot}}) + C_{12}(P_{\text{tot}}) - \frac{2C_{12}^2(P_{\text{tot}})}{C_{11}(P_{\text{tot}})}\right] + P\left[1 - \frac{C_{12}(P_{\text{tot}})}{C_{11}(P_{\text{tot}})}\right]\right\}. \tag{13}$$

It is clear from Fig. 1 that the effect of nonlinear elasticity originating from the pressure dependence of C_{ij} leads to decrease in dE_E/dP in InGaAs/GaAs QWs. The reduction in dE_E/dP in this case is stronger than in the case when third-order elasticity is taken into account. However, both theoretical results are in reasonable agreement with the experimental data.

Now, let us focus on dE_E/dP in (001)-oriented zinc-blende InGaN/GaN QWs. There is only one experimental study of dE_E/dP in these structures. In Ref. 15, the photoluminescence measurements of $E_E(P)$ were carried out for three $In_{0.1}Ga_{0.9}N/GaN$ QWs with different QW widths. The values of dE_E/dP were obtained from the linear fits to measured dependences of $E_E(P)$ in the range of P from

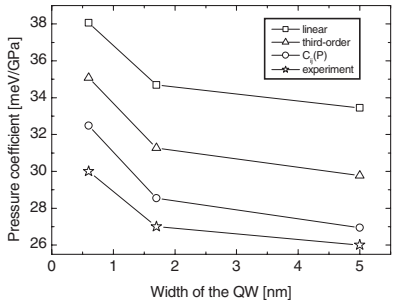

FIG. 2. Values of dE_E/dP for $In_{0.1}Ga_{0.9}N/GaN$ QWs as functions of QW width. Stars represent the experimental results taken from Ref. 15. Squares, triangles, and circles correspond to the theoretical results which are obtained using (i) the linear theory of elasticity, (ii) the third-order elasticity theory, and (iii) nonlinear elasticity originating from the pressure dependence of the second-order elastic constants, respectively. Solid lines are added to guide the eyes.

0 to 7 GPa.[15] To be consistent with the experimental procedure used in Ref. 15, we determine dE_E/dP using the linear regression for calculated dependences $E_E(P)$ in the corresponding range of pressures. In Fig. 2, we compare the experimental data taken from Ref. 15 with the results of calculations obtained using (i) the linear theory of elasticity, (ii) the third-order elasticity theory, and (iii) nonlinear elasticity arising from the pressure-dependent second-order elastic constants. Again, one can see that the usage of third-order elasticity leads to significant decrease in dE_E/dP, which significantly improves the agreement between theoretical and experimental results. However, better agreement between the theoretical and experimental values of dE_E/dP is obtained when instead of third-order elasticity, the pressure dependence of the second-order elastic constants is taken into account.

In conclusion, we have demonstrated that for strained (001)-oriented InGaAs/GaAs and InGaN/GaN QWs, the values of dE_E/dP calculated using the third-order elasticity theory are significantly lower than the results obtained by using the linear theory of elasticity. The usage of third-order elasticity improves considerably the description of the pressure tuning of strains in the QWs which results in better agreement between the theoretical and experimental values of dE_E/dP in the considered structures.

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