Theory of Piezoelectric Fields in InGaAs Site-Controlled Quantum Dots

Sorcha B. Healy\textsuperscript{1} and Eoin P. O'Reilly\textsuperscript{1,2}
\textsuperscript{1}Tyndall National Institute, Lee Maltings, Cork, Ireland
\textsuperscript{2}Department of Physics, University College Cork, Cork, Ireland
eoin.oreilly@tyndall.ie

Abstract. We present a surface integral method to calculate the piezoelectric polarisation potential due to a strained quantum dot (QD) grown on a (111)-oriented substrate. We show that the potential is reduced in a QD compared to a strained quantum well of the same height, but that there still remains a significant polarisation field across (111)-oriented InGaAs/GaAs QD structures. We use the 8-band $k.p$ method to show that these piezoelectric fields must be included to explain the measured interband transition energies in a series of site-controlled QDs with increasing height and In composition.

1. Introduction

Most interest in InGaAs/GaAs quantum dots (QDs) has to date focused on dots grown on an (001) substrate. Such dots experience a small built-in piezoelectric potential, due to the shear components of strain, $\varepsilon_{ij}$ ($i \neq j$). It is well established that there can be a large piezoelectric polarisation, due to the shear strain, across a strained quantum well structure grown on a (111) substrate [1]. A large piezoelectric potential may also be expected in (111)-oriented quantum dots. Very high quality growth has recently been demonstrated of InGaAs/GaAs site-controlled QDs, grown by metal-organic vapour phase epitaxy on GaAs (111)B substrates, pre-patterned with inverted pyramids [2]. These dots offer both high uniformity and spectral purity. The inhomogeneous broadening of excitonic emission from an ensemble of QDs was found to be unusually narrow, with a standard deviation of 1.19 meV and the spectral purity of emission lines from individual dots was found to be very high (18–30 µeV). It is therefore timely to undertake a theoretical analysis of the electronic structure of such dots. We investigate here the role of the built-in piezoelectric potential in such structures. We present a surface integral method to determine the polarisation potential, which we then use to estimate the potential variation in (111)-oriented strained QDs. We then use the 8-band $k.p$ method to calculate the effect of the piezoelectric potential in site-controlled QDs, showing that the built-in polarisation field has only a small effect in thin QDs with low In composition, but becomes of increasing importance, and must be taken into account, as the In composition and dot height are increased.

2. Derivation of Polarisation Potential in a (111)-oriented Quantum Dot

We derive here a semi-analytical expression to estimate the built-in polarisation potential in a (111)-oriented quantum dot. We follow the approach used by Davies [3]. This approach has been used previously to identify the key features of the piezoelectric potential in (001)-oriented QDs [3].
providing results which reproduce well the trends found in more realistic atomic calculations of both strain and piezoelectricity. The method omits any nonlinear dependence of the piezoelectric coefficients. Such terms are expected to be more important with increasing strain and In composition [4]. We expect, because we are considering dots with relatively low indium composition and strain, that the linear terms are still of greatest importance in the dots considered here. Davies showed that the polarisation potential $\phi(r)$ at $r = (x,y,z)$ can be given by an integral over the volume of the dot for a strained dot of constant composition buried in an infinite matrix:

$$\phi(r) = -C \int_{\text{Dot}} f(r, r') d^3r'$$

where $e_{ij}$ is the piezoelectric coefficient, $\varepsilon_0$ is the dielectric constant, $\varepsilon_s$ is Poisson’s ratio. The calculations assume that the elastic behaviour is linear and isotropic, with the same elastic constants everywhere, and also that $e_{ij}$ and $\varepsilon_s$ have the same value in the dot and the matrix. Using the divergence theorem, the volume integral in Eq. (1) can be brought to the surface integral

$$\Phi = \int_{V_G} \nabla \cdot \mathbf{\varepsilon} \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} = \int_{S_G} \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} \cdot \mathbf{\varepsilon} \cdot \mathbf{\varepsilon}$$

Davies introduced a number of functions $G(r, r')$ which satisfy this requirement, the simplest for a (001)-oriented dot being of the form $G(r, r') = G_1(r, r') \gamma$. This form of $G$ is very useful to analyse dots grown on a (001) substrate, because it allows the piezoelectric potential to be evaluated by integrating over the (001)-oriented dot faces. We now derive a form of $G$ suitable to analyse QDs grown on a (111) substrate.

We first rotate the basis vectors so that $x_3$ points along the (111) direction, with the new unit vectors defined in terms of the original Cartesian coordinates as

$$\hat{x}_i = \frac{1}{\sqrt{6}}(1,1,-2); \quad \hat{x}_2 = \frac{1}{\sqrt{2}}(-1,1,0); \quad \hat{x}_3 = \frac{1}{\sqrt{3}}(1,1,1)$$

When we transform the integrand in Eq. (1) to be a function of the rotated coordinates, we obtain a large and unwieldy form for $f(r, r')$. It is however possible to then find a function $G_i(r, r') \hat{x}_i$ such that

$$\frac{\partial G_i}{\partial x_3} = f(r, r')$$

with the potential given by $\phi(r) = C[I_1 + 5I_2 + I_3]$, where $C$ is given in Eq. (1), and

$$I_1 = \int \left( (x_i - x_i') \hat{n}_i dS' \right); \quad I_2 = \int \hat{n}_i dS'; \quad I_3 = \int \frac{\sqrt{2}(x_i - x_i') [3(\rho - \rho') + 2(x_i - x_i')^2 \cos(\theta)] \hat{n}_i dS'}{\rho - \rho'}$$

where $\rho = (x_1, x_2)$ and $\theta$ is the angle between $\rho - \rho'$ and the $x_3$ axis. At $x_1 = x_2 = 0$ (i.e. along the central $x_3$ axis of the dot) $I_1$ will integrate to zero if the dot is of sufficiently high symmetry. For a cylinder, truncated cone or indeed triangular pyramid, this is the case.

We have shown previously when studying the built-in potential in III-N wurtzite dots that the integrals $I_1$ and $I_2$ have analytical solutions along the central axis $(x_1 = x_2 = 0)$ for a wide range of dot shapes [5]. We therefore use Eq. (3) to compare the piezoelectric potential in a (111)-oriented QD with that in a (111)-oriented quantum well (QW) of the same height. We consider the piezoelectric potential in a cylindrical QD of height $h$ and radius $R$ as a function of $f = h/R$, where $f = 0$ for a QW and $f = 0.04 - 0.25$ for the site-controlled QDs which we consider below. The potential drop $\phi_{\text{max}}^\alpha$ across a (111)-oriented strained QW of height $h$ is given from Eq.(3) as

$$\phi_{\text{max}}^\alpha = \frac{2e_{ij} h \varepsilon_0}{\sqrt{3} e_s \varepsilon} \frac{1 + \nu}{1 - \nu}$$

We can solve Eq.(3) for finite $R$ to show that the piezoelectric potential drop $\phi_{\text{max}}^\alpha$ between the top and bottom surface of an isolated cylindrical dot will vary with $f$ for small $f$ as
\[ \varphi_{\text{max}} = \varphi_{\text{max}}^0 \left( 1 - \frac{9}{8} f \right) \]

The solid lines with symbols in Figure 1 show the calculated variation of peak-to-peak potential as a function of dot radius, \( R \), for a dot height \( h \) of 2 nm and of 8 nm, along with a fit to the calculated variation using Eq. (5). It can be seen that Eq. (5) is within 5% of the calculated value until \( f = 0.36 \) (\( R = 22 \) nm for \( h = 8 \) nm in Fig. 1). Two factors modify the strain-induced polarization potential in a QD: first, the effect of the finite dot size, which reduces the size of the region in which there is a piezoelectric-induced polarization, and second the effects of the change in strain distribution in the QD compared to a QW of the same height. \( \varphi_{\text{max}} \) would vary only as \( \varphi_{\text{max}}^0 (1 - 0.5 f) \) if the strain distribution was identical in the dot and in the well. This is a much weaker variation than that given in Eq. 5, showing that the strain redistribution in the dot compared to the well makes a significant contribution to the overall reduction in the piezoelectric potential in the dot.

**Figure 1:** Calculated variation with radius of potential drop across a cylindrical dot of height 2 nm (black line) and 8 nm (green lines) compared with the analytical expression of Eq. (5).

Fig. 2 shows the calculated variation of the polarization potential across an 8nm In\textsubscript{35}Ga\textsubscript{65}As/GaAs QW, as well as the potential along the \( x_3 \) axis through the centre of a cylinder of the same height, and radius \( R = 22 \) nm, and for a 8 nm high truncated cone with radius \( R_t = 22 \) nm and \( R_b = 18.9 \) nm on the top and bottom surfaces respectively. The calculations in Fig. 2 assume a linear variation of piezoelectric coefficient \( e_{14} \) with In concentration. The reduction in potential difference across the dots compared to that across a QW of the same height and composition can be clearly observed. Nevertheless, we still calculate a potential drop of order 200 meV, equivalent to a built-in field of 2.5 \( \times 10^5 \) V cm\(^{-1}\) across the QDs. We show below that polarisation fields of this scale can have a significant impact on the ground state transition energy in the QD.

The inset in Fig. 2 shows the potential variation over a larger distance along the \( x_3 \) axis of the conical QD. We see that the potential outside the QD returns towards zero, and can even change sign at larger distances from the dot. This is to be contrasted with the QW, where the potential remains constant outside the well. The change in sign of the potential at large \( x_3 \) values arises due to the strain distribution in the matrix material surrounding the dot. Because the dot expands along the \( x_3 \) axis, the surrounding matrix material must shrink along this direction, thus accounting for the observed variation in polarisation field and potential.

### 3. Transition Energies in Site-controlled InGaAs QDs

We have developed an 8-band \( k.p \) model to calculate the electronic structure in (111)-oriented QD structures. Details of the method can be found in [6]. The initial calculations which we presented using this method did not include the piezoelectric potential [7]. The results of these calculations were...
compared with measurements of the ground state transition energy in a series of site-controlled InGaAs/GaAs QDs with increasing In composition and nominal dot height. The nominal growth rate is lower than the actual growth rate for a site-controlled QD, with the dot growing at a faster rate than the surrounding side walls. We found good agreement in our initial calculations between the measured transition energies in a series of dots with nominal height of 0.5nm when we assumed the actual dot growth rate to be ~2.5 times faster than the nominal rate. However our calculations, without the piezoelectric potential, overestimated the transition energies for taller dots.

Figure 3 compares the calculated (open symbols) and measured transition energies (solid symbols) for dots of various In compositions and heights, with the piezoelectric potential included in the calculations. The previous calculations underestimated by about 50 meV the transition energy of 35% In dots with nominal height of 2 nm. It can be seen that there is now good agreement between theory and experiment across the full composition and height range considered, confirming the importance of including piezoelectric effects in (111)-oriented InGaAs QDs with higher In composition. The dots were prepared for measurement in two different ways: either by surface-etching or back-etching. A systematic reduction in transition energy was observed in the back-etched samples [2], so the back-etched samples in Fig. 3 have all been shifted upwards in energy to account for this processing-induced shift in emission energy.

![Figure 3: Comparison of theoretical (pen symbols) and experimental (filled symbols) interband transition energies for inverted pyramidal site-controlled dots of varying heights and In concentration.](image)

**4. Conclusions**

In summary, we presented a simple analytical framework to estimate the size of the piezoelectric potential in (111)-oriented strained quantum dots. We showed that although the field is reduced compared to the value in a quantum well of the same composition it can still significantly influence the transition energy in dots with higher In composition, in agreement with the results of recent experimental measurements on a series of (111)-oriented site-controlled InGaAs/GaAs QD structures.

**Acknowledgements** This work was supported by Science Foundation Ireland. We thank Stefan Schulz, Emanuele Pelucchi and Paudie Scanlon for useful discussions.

**References**

[1] D. L. Smith and C. Mailhiot, Rev. Mod. Phys. 62, 173 (1990)
[2] L. O. Mereni, V. Dimastrodonato, R. J. Young, E. Pelucchi, Appl.Phys.Lett. 94, 223121 (2009)
[3] J.H. Davies, J. Appl. Phys. 84, 1358 (1998)
[4] G. Bester, X. Wu, D. Vanderbilt and A. Zunger, Phys.Rev. Lett. 96, 187602 (2006)
[5] D.P. Williams, A.D. Andreev, E.P. O’Reilly and D.A. Faux, Phys. Rev. B. 72, 235318 (2005)
[6] S.B. Healy and E.P. O’Reilly, IEEE J. Quantum Electron. 46, 742 (2010)
[7] S. B. Healy et al., Physica E (to be published; doi:10.1016/j.physe.2009.11.099)