Phonons in strained semiconductor nanostructures

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Abstract. Semiconductor nanostructures contain significant strain contributions, which in turn influence phonon properties. Model calculations for spherically shaped nanodots embedded in a host crystal are presented using the Keating model as interaction potential. It is shown that the local dot phonon density of states is to a good approximation given by the phonon density of the bulk material under identical strain. We present calculations of effective elastic constants for various strain situations employing ab initio density functional theory.

1. Introduction
If different semiconductor compounds should be combined for electronic or optical device applications then not only the band gap but also the lattice constants vary in general. Epitaxial growth techniques enable to accomodate this lattice mismatch in a strain field without structural defects. The strain field itself can be utilized in optimizing the desired device properties, since the electronic properties couple to it e.g. by deformation potentials. On the other hand, also the vibrational modes are changed. The interatomic distances and therefore the local force constants become spatially dependent in inhomogeneous nanostructures.

The theoretical description of strain relaxation and phonons can be done within the framework of continuum elasticity or atomistic models. Major obstacles for theoretical treatment are the large number of atoms involved and missing suitable interaction potentials. Although there is an agreement on second order elastic constants for most semiconductors, there are still discrepancies already for the next higher order. Restricting to small displacements considered for acoustic phonons allows the treatment by linear elasticity or equivalently by harmonic approximations in the interaction potentials. The relevant input parameters, which are effective elastic constants or the coupling force constants, need to include the structural relaxation due to strain and are in this case of existing inhomogeneities spatially dependent.

2. Theory
The governing equation for acoustic phonons with frequency $\omega_\nu$ in continuum elasticity theory is given by

$$-\omega_\nu^2 \rho(\mathbf{r}) u_i(\mathbf{r}) = \sum_j \frac{\partial}{\partial x_j} \sigma_{ij}(\mathbf{r}).$$

The mass density $\rho(\mathbf{r})$, the displacement $\mathbf{u}(\mathbf{r})$, and the stress tensor $\mathbf{\sigma}$ are in general spatially dependent. Within linear elasticity the stress tensor is given by second order elastic constants.
$C_{ijkl}$ and the strain tensor $\varepsilon$ (Hooke’s law)

$$\sigma_{ij} = \sum_{kl} C_{ijkl} \varepsilon_{kl}. \quad (2)$$

If the material is under strain the elastic constants have to be replaced by effective ones. If the local strain lowers the symmetry the number of independent elastic constants $C_{ijkl}$ increases (see below) and their numerical values will depend on the amount of local strain. Under a local assumption they can be calculated for homogeneous deformations of the unit cell by density functional theory (DFT) as done here with the ABINIT program [1] or deduced by continuum elasticity [2]. A direct calculation is desired, since the lattice mismatch in semiconductor nanostructures can reach values exceeding even the elasticity limits including third order elastic constants [3].

3. Molecular dynamics simulations

To test a local assumption we carried out molecular dynamics simulations, which allows to calculate the phonon density of states (DOS)

$$G(\omega) = \frac{1}{3N} \sum_\nu \delta (\omega - \omega_\nu), \quad (3)$$

for nanostructures. Solving Newton’s equation of motion with an initial velocity $v_{i,\alpha}(t = 0)$ for atom $i$ of species $\alpha$ and forming the velocity autocorrelation function $Z_\alpha(t)$

$$Z_\alpha(t) = \frac{\langle v_{i\alpha}(0) \cdot v_{i\alpha}(t) \rangle}{\langle v_{i\alpha}^2(0) \rangle}, \quad (4)$$

enables to deduce the density of states via Fourier transformation

$$G(\omega) = \frac{1}{N} \sum_\alpha \frac{2N_\alpha}{\pi} \int_0^\infty Z_\alpha(t) \cos(\omega t) dt. \quad (5)$$

The initial conditions have to be chosen such that the atoms are in their equilibrium position and start with an uncorrelated velocity distribution (e.g. box distribution) with a chemically specific variance $\langle v_k^2(0) \rangle = b^2/M_k$, where $b$ is a constant and $M_k$ is the mass of the atomic species $k$. The simulations are carried out using a damped Verlet algorithm for the structural relaxation and the velocity Verlet for the lattice dynamics. Since the investigated model system is a small inclusion in an extended host crystal, the density of states is mainly determined by the host crystal properties. Therefore we investigate the local density of states limiting the summation in Eq. 5 to the atoms inside the dot.

Since we are interested in more general properties we have chosen a valence-force field interaction of the Keating type [4], which is especially suited for tetrahedrally bonded semiconductors with cubic symmetry. Its advantage is the already existing parametrization for a large variety of semiconductors. The two Keating parameters $\alpha$ and $\beta$ can directly be determined from the elastic constants $C_{11}$ and $C_{12}$ and the lattice constants. However, it is known that this model lacks in some quantitative details: Most of the third order elastic constants are zero, except $C_{144}$ and $C_{166}$ which are too small [5], and the second order constant $C_{44}$ is underestimated for compound III-V and II-VI semiconductors.
4. Results and Discussion
The structural model we investigate is a spherically shaped InAs-dot in a GaAs host crystal. Numerical parameters are taken from the Keating paper [4]. First the relaxed atomic positions and displacements are determined, followed by the calculation of the strain field. Results for the dilation, i.e. $\text{tr}(\epsilon)$, are shown in Fig. 1 left and the non-diagonal element $\epsilon_{zx}$ right. The strain inside the dot is nearly constant and is approximately pure hydrostatically, in qualitative agreement with isotropic continuum elasticity. Secondly, the MD simulation is used to calculate the local dot phonon density as described above. The local dot phonon density is plotted in Fig. 3. Comparing this local phonon density now with the phonon density of a homogeneously, by 3.18% hydrostatically compressed (identical to the dilation inside the dot of Fig. 1) InAs crystal, we find surprisingly an overall perfect match. Although we presented only a single, very special result, it motivates an approach to be taken in further studies. Treating the spatially varying strain as being constant locally, therefore neglecting gradient terms, effective elastic
constants or force constants can be calculated directly from DFT.

We present calculations of effective elastic constants for strained units cells in order to investigate the possible local variations of the elastic constants to be used in Eq. (1). Calculations are carried out with the ABINIT simulations package [1] within local density approximation (LDA) and within the pseudopotential approach [6]. Results for hydrostatic strain can be found elsewhere [3]. As examples we show Fig. 3 results for GaAs under biaxial strain without (left) and with volume conservation (right). The values at \( \alpha = 0 \) agree very well with data from recent literature (theory [7] and experiment [8]). Recent calculations employing the general gradient approximation showed larger discrepancies [9]. The reduced symmetry of the unit cell leads to differences between the elastic constants \( C_{11} = C_{22} \) and \( C_{33} \) as well as \( C_{13} = C_{23} \) and \( C_{12} \), which are identical for cubic symmetry (\( \alpha = 0 \)).

![Figure 3. Effective elastic constants for GaAs under biaxial strain without (left \( \epsilon = (\alpha + \alpha^2/2)(1,1,0,0,0,0) \) in Voigt’s notation) and with volume conservation (right \( \epsilon = (\alpha + \alpha^2/2)(1,1,-2,0,0,0) \). The value \( \alpha = 0 \) corresponds to the equilibrium lattice constant.]

5. Summary
Phonons in semiconductor nanostructures are influenced by strain, which changes locally the effective elastic properties. We presented molecular dynamics calculations for the phonon density of spherical nanodots. The local dot phonon density agrees well with the density of an equally strained bulk crystal. Therefore, it seems to be a good approximation in phonon calculations to treat the elastic constants or force constants, respectively, dependent on the local strain value. The parametrization can be done through DFT calculations of effective elastic constants for various strain situations.

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