

Numerical simulation of the longitudinal
dispersion bands of phonons in a
 $(GaAs)_1/(AlAs)_1$ superlattice along the $[001]$
direction

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Abstract

The longitudinal vibrational properties of an “ultra-thin” $(GaAs)_1/(AlAs)_1$ superlattice are studied along the $[001]$ direction using a series of approximations, so that the problem is reduced to the study of a simple linear chain of harmonic oscillators.

Phononic bands are calculated numerically, along with displacements in Γ and the frequency of $GaAs/AlAs$ -like LO modes, both in the nearest neighbour and in the next-nearest neighbour approximation. Comparison is made with experiments and more refined theoretical models.

This work shows how even simple models are able to capture the essential features of the physical problem, such as correctly hinting at the presence of confined modes.

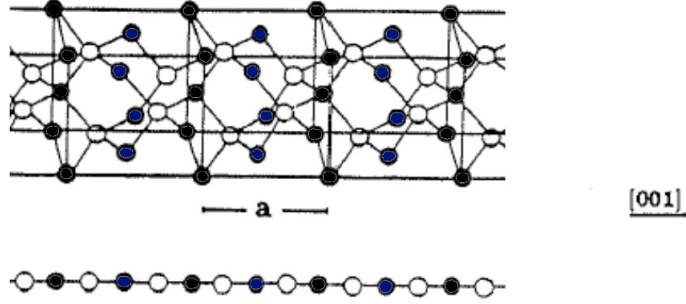


Figure 1: The superlattice used in this study and its projection on the $[001]$ direction. Gallium atoms are painted black, Aluminium ones are blue and Arsenic is white. Note that, when projecting on the $[001]$ direction, each of the planes can be treated as a single atom.

Introduction

The aim of this work is to study the vibrational properties of a superlattice in a particularly simplified model, to show how even simple models lead to a rather good level of agreement with both more sophisticated theoretical models and experiments.

Normal vibrational modes of a quantized elastic system are called phonons. They are collective excitations - i.e. bosonic quasiparticles - and their dispersion relations are related to a number of properties of the system itself.

A superlattice is a kind of metamaterial made up from two (or more) materials, in a periodic structure. An ideal $(GaAs)_1/(AlAs)_1$ superlattice is made up from one layer of $GaAs$ followed by one of $AlAs$ in a periodic matter (see Figure 1).

In general, the vibrational properties of a crystalline solid can be studied through lattice dynamics; thanks to the periodicity of the system the problem can be further reduced to the solution of the dynamics of the atoms in the unitary cell.

Three more approximations will be employed:

1. **The Born-Oppenheimer approximation**, where ionic and electronic motions can be treated independently.
2. **The harmonic approximation**, which consider the atomic cores as simple harmonic oscillators.
3. **The nearest neighbour approximation**, where for each atom the interaction between anything other than the closest atoms next to it are neglected.

The third will later be relaxed to include also the next to nearest neighbour. Thanks to these approximation it is possible to reduce the quanto-mechanical problem of vibrations in a solid to the classical one of finding the normal modes

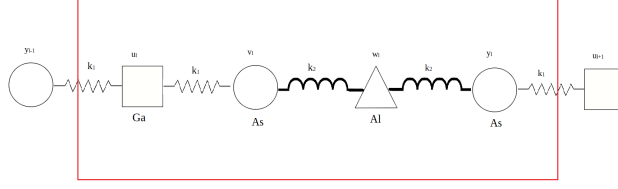


Figure 2: The 4-atom unitary cell analysed in this work in the nearest neighbours approximation.

k_1 is the elastic constant between Ga and As and k_2 is the elastic constant between Al and As. Considering an infinite linear chain, u_l represents the displacement of the l -th (arbitrary origin) atom of Ga, v_l the displacement of the l -th atom of As of the GaAs molecule, w_l the displacement of the l -th atom of Al, and y_l the displacement of the l -th atom of As of the AlAs molecule.

of a linear chain of harmonic oscillators. The dispersion bands (how the frequency depends on \vec{q} , the wavevector of the mode) can then be found through a simple numerical algorithm.

Only the [001] direction will be considered, so that the materials' layers follow each other orthogonally with respect to this direction. Only longitudinal vibrations have been taken into account.

Physical Problem

Because of the approximation used in this study, the relevant physical model is a linear chain of atoms in which the unit cell is made up of 4 atoms, Ga-As-Al-As as in Figure 2, and - at least in first approximation - only interaction with nearest neighbour needs to be taken into account.

The following system results (see Figure 2 for the meaning of each symbol):

$$\begin{cases} M_{Ga}\ddot{u}_l = -k_1(u_l - v_l) - k_1(u_l - y_{l-1}) \\ M_{As}\ddot{v}_l = -k_1(v_l - u_l) - k_2(v_l - w_l) \\ M_{Al}\ddot{w}_l = -k_2(w_l - v_l) - k_2(w_l - y_l) \\ M_{As}\ddot{y}_l = -k_2(y_l - w_l) - k_1(y_l - u_{l+1}) \end{cases} \quad (1)$$

To solve this system we impose that every displacement has the form of a plane wave:

$$\begin{cases} u_l = Ue^{i(qla - \omega t)} \\ v_l = Ve^{i(qla - \omega t)} \\ w_l = We^{i(qla - \omega t)} \\ y_l = Ye^{i(qla - \omega t)} \end{cases} \quad (2)$$

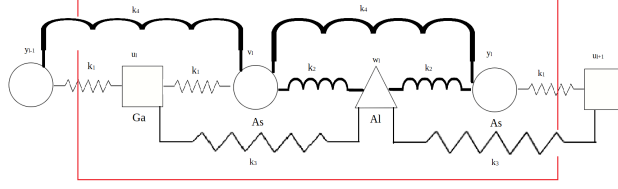


Figure 3: The 4-atom unitary cell analysed in this work in the next-nearest neighbours approximation.

k_1 is the elastic constant between Ga and As and k_2 is the elastic constant between Al and As, while k_3 and k_4 are the cation-cation and anion-anion force constants, respectively. Considering an infinite linear chain, u_l represents the displacement of the l -th (arbitrary origin) atom of Ga, v_l the displacement of the l -th atom of As of the GaAs molecule, w_l the displacement of the l -th atom of Al, and y_l the displacement of the l -th atom of As of the AlAs molecule.

where a is the lattice parameter of the superlattice (sum of the lengths of the respective unit cells). Solving the system for ω and then varying \vec{q} will lead to the dispersion bands; since the unit cell has 4 atoms the system to solve is 4-dimensional and so 4 dispersion bands are expected.

Similarly, one can obtain the following system when in the next-nearest neighbour approximation (see Figure 3):

$$\begin{cases} M_{Ga}\ddot{u}_l = -k_1(u_l - v_l) - k_1(u_l - y_{l-1}) - k_3(u_l - w_{l-1}) - k_3(u_l - w_l) \\ M_{As}\ddot{v}_l = -k_1(v_l - u_l) - k_2(v_l - w_l) - k_4(v_l - y_{l-1}) - k_4(v_l - y_l) \\ M_{Al}\ddot{w}_l = -k_2(w_l - v_l) - k_2(w_l - y_l) - k_3(w_l - u_l) - k_3(w_l - u_{l+1}) \\ M_{As}\ddot{y}_l = -k_2(y_l - w_l) - k_1(y_l - u_{l+1}) - k_4(y_l - v_l) - k_4(y_l - v_{l+1}) \end{cases} \quad (3)$$

Values of the force constants are taken from *ab initio* calculations [1, Table 2], assuming that they are the same as the single lattice case. Masses, on the other hand, are taken from [2]. Since the force constants k_1 and k_2 are so similar (equal up to less than 5%), their difference can be neglected, substituting their average value for both of them. This is sometimes called the “Mass approximation”, since only the mass difference between the two atoms is taken into account. Of course, neglecting also the mass difference would take the problem back to a simple lattice instead of a superlattice.

As for the next-nearest neighbour case, $k_3(k_4)$ is taken to be the average of the anion-anion(cation-cation) constant of GaAs and AlAs.

Numerical solution

The problem of finding the dispersion bands of *GaAs/AlAs* reduces - in the nearest neighbour approximation - to the solution of the system showed in Equation 3.

Substituting the plain waves for the displacements in Equation 2, after some algebra one gets:

$$\begin{cases} (-\omega^2 M_{Ga} + 2k_1)U - k_1 V - (k_1 e^{-iqa})Y = 0 \\ -k_1 U - k_2 W + (-\omega^2 M_{As} + k_1 + k_2)V = 0 \\ (-\omega^2 M_{Al} + 2k_2)W - k_2 V - k_2 Y = 0 \\ -(k_1 e^{iqa})U - k_2 W + (-\omega^2 M_{As} + k_1 + k_2)Y = 0 \end{cases} \quad (4)$$

This is convenient as the system's associated matrix is already written in the form $[-\omega^2 I + A]$, where

$$A = \begin{pmatrix} \frac{2k_1}{M_{Ga}} & -\frac{k_1}{M_{Ga}} & 0 & -\frac{k_1}{M_{Ga}}e^{-iqa} \\ -\frac{k_1}{M_{As}} & \frac{k_1}{M_{As}} + \frac{k_2}{M_{As}} & -\frac{k_2}{M_{As}} & 0 \\ 0 & -\frac{k_2}{M_{Al}} & \frac{2k_2}{M_{Al}} & -\frac{k_2}{M_{Al}} \\ -\frac{k_1}{M_{As}}e^{iqa} & 0 & -\frac{k_2}{M_{As}} & \frac{k_1}{M_{As}} + \frac{k_2}{M_{As}} \end{pmatrix} \quad (5)$$

so it is possible to solve the secular equation and find ω^2 . Varying q inside the first Brillouin Zone (the direction is fixed so only the magnitude varies) one arrives at the dispersion relation, whereas the eigenvectors of A are the displacement of the single atoms in the chain.

Similarly, for next-nearest neighbour:

$$A' = \begin{pmatrix} \frac{2(k_1+k_3)}{M_{Ga}} & -\frac{k_1}{M_{Ga}} & -\frac{k_3(1+e^{-iqa})}{M_{Ga}} & -\frac{k_1}{M_{Ga}}e^{-iqa} \\ -\frac{k_1}{M_{As}} & \frac{k_1+k_2+2k_4}{M_{As}} & -\frac{k_2}{M_{As}} & -\frac{k_4(1+e^{-iqa})}{M_{As}} \\ \frac{k_3(1+e^{iqa})}{M_{Al}} & -\frac{k_2}{M_{Al}} & \frac{2(k_2+k_3)}{M_{Al}} & -\frac{k_2}{M_{Al}} \\ -\frac{k_1}{M_{As}}e^{iqa} & -\frac{k_4(1+e^{iqa})}{M_{As}} & -\frac{k_2}{M_{As}} & \frac{k_1+k_2+2k_4}{M_{As}} \end{pmatrix} \quad (6)$$

To obtain the solution of the problem a numerical approach was undertaken: a program in C++ has been written which, after discretizing the Brillouin Zone in a selected number of points, calculates the appropriate wavevector q for each points, and then fills the matrix in Equation 6 with the appropriate values. Finally, using a subroutine of the *Eigen* linear algebra package, it finds eigenvectors and eigenvalues for each matrix.

The result is a discretized version of the dispersion relation, which is then plotted using GNUPLOT.

Results and Comparison

In Figure 4 one can see the band structure as calculated using the Mass and nearest neighbours approximations and using only the nearest neighbours ap-

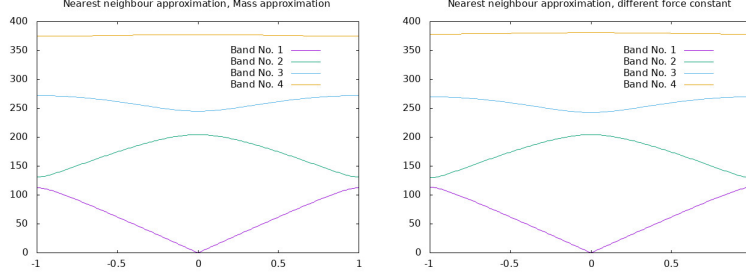


Figure 4: Calculated band structure in the $[001]$ direction.

Left: using nearest neighbour and Mass approximations. **Right:** using only nearest neighbour approximation. It's evident how similar the two results are, justifying the Mass approximation.

Band 1 shows a linear dispersion, as expected from an acoustic mode, whereas band 3 and 4 are those that describe GaAs and AlAs-like LO modes, respectively.

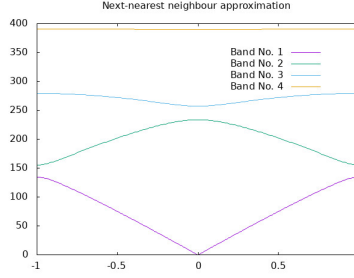


Figure 5: Calculated band structure in the $[001]$ direction, using next-nearest neighbour approximations. It appears as an upward shifted version of the nearest neighbour one, but also features reduced band-gaps.

proximation, while the results of the next-nearest neighbour approximation is shown in Figure 5. Figure 6 shows the amplitude of the normal modes in Γ .

The results whose comparison to known literature is easier is the frequency of the GaAs(AlAs)-like modes; this is because these frequencies can be obtained from Raman spectroscopy and in part because their interpretation had a certain degree of controversy when they were first observed [1]. Direct comparison with ([1, Fig.6],[3, 4]) can be done, to see how the present work stacks up with theoretical (from linear response DFT) and experimental values found in literature. Comparison can be seen in Table 1.

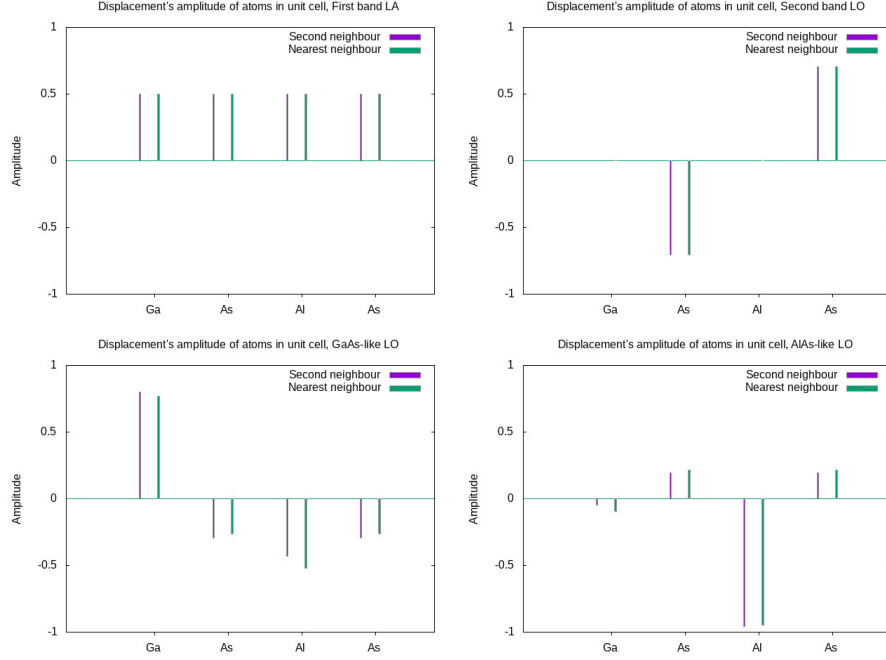


Figure 6: Calculated amplitudes (*nearest neighbour* and *next-nearest neighbour*) of the four bands, in the high symmetry point Γ .

The first mode is appropriately a rigid translation, since it's a zero frequency mode. The third and fourth mode should respectively be the GaAs-like and AlAs-like LO mode. While the AlAs-like mode does indeed display some degree of confinement, no such thing can be said for the GaAs-like mode, showing the limitation of this simple model.

	Present Work (NN)	Present Work (SN)	Theory [1]	Experiment [3, 4]
GaAs-like	245	257	266	275
AsAl-like	377	389	399	385

Table 1: Comparison of LO modes frequency. The first two columns are values calculated in the present work, with Nearest-Neighbour (NN) and Next-Nearest Neighbour approximation, whereas the last two columns show the values calculated in [1] using DFT and values obtained from Raman spectroscopy in [3, 4], respectively. All values in cm^{-1} .

Conclusions

A simple model has been used to calculate the vibrational properties of an “ultra-thin” $(GaAs)_1/(AlAs)_1$ superlattice as a linear chain of atoms oscillating along the $[001]$ direction. Dispersion relations have been plotted, showing correctly the linear acoustic band and the flat $AlAs$ -like mode.

Comparison of the calculated $GaAs/AlAs$ -like frequencies to both experiment and more sophisticated theoretical models gives a satisfactory level of agreement, given the crudeness of the model. In particular the $AlAs$ -like mode frequency is calculated with a discrepancy of $\sim 5\text{ cm}^{-1}$ from the experiment (better than the one reached by the DFT calculation), while the $GaAs$ -like mode shows a more significant discrepancy ($\sim 20\text{ cm}^{-1}$).

On the other hand, confinement is only partially exhibited by this model, as shown by looking at the amplitudes of the normal modes.

References

- [1] E.Molinari, S.Baroni, P.Giannozzi, S. de Gironcoli (1991). *Phonon spectra of ultrathin GaAs/AlAs superlattices: An ab initio calculation*. **Light Scattering in Semiconductor Structures and Superlattices (pp 39-52)**
- [2] Juris Meija *et al.* (2016). "Atomic weights of the elements 2013 (IUPAC Technical Report)". **Pure Appl. Chem. 88 (3): 265–291.**
- [3] M.Cardona, T.Suemoto, N.E. Christensen, T.Isu, K.Ploog (1987) *Electronic and vibronic structure of the (GaAs)₁(AlAs)₁ superlattice*. **PRB 36, 5906**
- [4] A.Ishibashi *et al.* (1986) *Raman scattering from (AlAs)(GaAs) ultrathin-layer superlattices*. **PRB 33 2887**