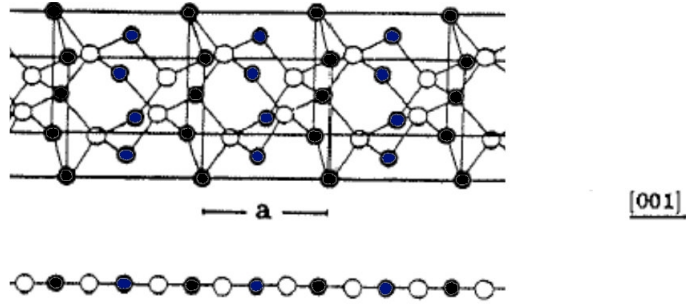


Numerical simulation of the longitudinal  
dispersion bands of phonons in a  
 $(GaAs)_1/(AlAs)_1$  superlattice

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February 3, 2020



**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.

## 1 Abstract

## 2 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 of quantized elastic system are called phonon. They are collective excitation - i.e. bosonic quasiparticles - and their dispersion relation are related to a number of properties of the system itself.

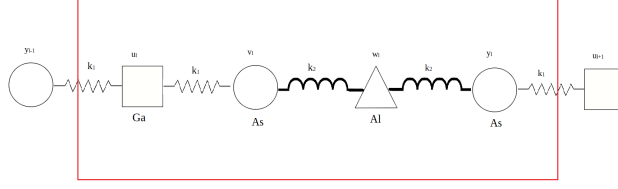
A superlattice is a kind of metamaterial made up from two (or more) materials, in a periodic structure. An ideal  $(GaAs)_1/(AlAs)_1$  superlattice is made up from one layer of  $GaAs$  followed by one of  $AlAs$  in a periodic matter (see Figure 1).

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

The second will later be relaxed to include also the next to nearest neighbour. Thanks to these approximation it is possible to reduce the quantum-mechanical



**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  $u_l$  represent 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,  $y_l$  the displacement of the  $l$ -th atom of As of the AlAs molecule.

problem of vibrations in a solid to the classical one of finding the normal modes. 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  $[0,0,1]$  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.

### 3 Physical Problem

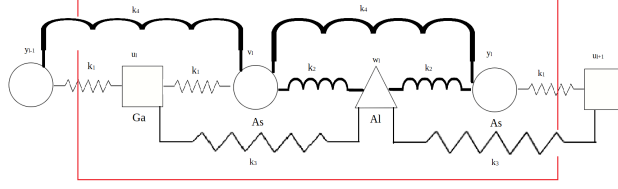
Because of the approximation used in this study, the relevant physical model is a linear chain of atoms in which the unit cell is made up by 4 atoms, Ga-As-Al-As as in Figure 2, and - at least in first approximation - only interaction with nearest neighbour needs to be taken into account.

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  $u_l$  represent 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,  $y_l$  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  $\omega$  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 2b], assuming that they are the same as the single lattice case. Masses, on the other hand, are taken from [2]. Since the force constant  $k_1$  and  $k_2$  are so similar (equal up to less than 5%), their difference can be neglected, substituting to both of them their average value. 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.

## 4 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 Equa-

tion 3.

Substituting into the system the solution for the plain wave 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  $\omega^2$ . 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:

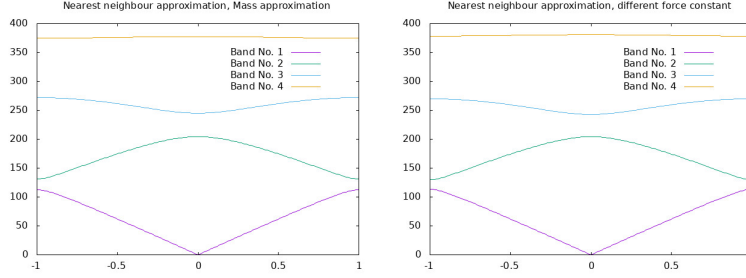
$$A' = \begin{pmatrix} \frac{2(k_1+k_3)}{M_{Ga}} & -\frac{k_1}{M_{Ga}} & -\frac{k_3(1+e^{-iqa})}{M_{Ga}} & -\frac{k_1}{M_{Ga}}e^{-iqa} \\ -\frac{k_1}{M_{As}} & \frac{k_1+k_2+2k_4}{M_{As}} & -\frac{k_2}{M_{As}} & -\frac{k_4(1+e^{-iqa})}{M_{As}} \\ \frac{k_3(1+e^{iqa})}{M_{Al}} & -\frac{k_2}{M_{Al}} &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

To obtain the solution of the problem a numerical approach was undertaken: a program in C++ has been written which, after discretising 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.

## 5 Results and Comparison

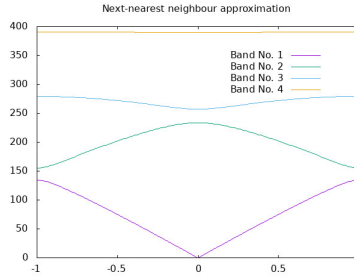
In Figure 4 one can see the band structure as calculated using the Mass and nearest neighbours approximations, only the nearest neighbours approximation, while the results of the next-nearest neighbour approximation is shown in Figure 5. [1, Fig ] shows the amplitude of the normal modes in  $\Gamma$ .



**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 3 and 4 are those that give rise to *GaAs* and *AlAs*-like 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.

The results whose comparison to known literature is easier is the frequency of the *GaAs*(*AlAs*)-like modes; this is because these frequency can be obtained from Raman spectroscopy. Direct comparison with ([1, Fig.6] and [3]) can be done, to see how the present works stacks up with theoretical(from linear response DFT) and experimental values found in literature. Comparison can be seen in ??.

## 6 Conclusion

A simple model has been used to calculate the vibrational properties of an “ultra-thin” *GaAs*/*AlAs* as a linear chain of atoms oscillating along the  $[001]$  direction.

Comparison of the *GaAs*/*AlAs*-like frequencies to both experiment and more sophisticated theoretical models gives a satisfactory level of agreement, given the

crudeness of the model. On the other hand, confinement is only partially exhibited by this model, as show 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)<sub>1</sub>(AlAs)<sub>1</sub> superlattice*. **PRL B 36, 5906**