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1996 J. Phys.: Condens. Matter 8 6329

(http://iopscience.iop.org/0953-8984/8/35/004)

J. Phys.: Condens. Matter **8** (1996) 6329–6336. Printed in the UK

Lattice dynamics of zinc-blende GaN and AlN: II. Superlattice phonons

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Received 2 January 1996, in final form 24 June 1996

Abstract. Phonon dispersions of (001)-oriented cubic GaN/AlN superlattices in the longitudinal polarization were calculated by a planar-force-constant model. Raman spectra were calculated by a bond-polarizability model. The confinement of the optical modes was discussed.

1. Introduction

Since the pioneering work of Esaki and Tsu [1] semiconductor superlattices (SLs) have received considerable attention due to their potential applications in devices. When two semiconductor constituents were put together to form SLs, novel electronic and vibrational properties appear which do not exist in the constituents. Great efforts have been paid to the growth, characterization, and application of semiconductor SLs.

In the preceding paper, the phonon dispersions of cubic GaN and AlN were studied by a Keating-type potential with the long-range Coulomb interactions taken into account [2]. The involved parameters were obtained by fitting them to the data transformed from the Raman experimental data of the wurtzite structures by a folding procedure. Because most of the SLs have been grown on (001)-oriented substrates, we will study the vibrational properties of cubic GaN/AlN (001) SLs in the present work, although SLs oriented in other directions can also be studied within the framework of the present theoretical model.

The paper is organized as follows. In section 2 the basic features of the planar-force-constant model and bond-polarizability model are described. In section 3 the calculated phonon dispersions and corresponding displacement patterns are given. The confinement of GaN-like and AlN-like modes is discussed. The conclusions are given in section 4.

2. Planar-force-constant model and bond-polarizability model

For (001)-oriented cubic GaN/AlN SLs, it can be shown by group theory that the longitudinal (L) and transverse (T) vibrations can be decoupled. This means that we can deal with the longitudinal and transverse vibrations separately. Then, a simple planar-force-constant model is used in the present work to calculate the phonon dispersions, rather than the three-dimensional calculations.

The sth nearest-neighbour planar-force-constant is denoted by k_s. For L vibrations we have $k_{-s} = k_s$ due to the symmetry condition. The dynamical matrix [3] for cubic GaN

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0953-8984/96/356329+08$19.50

or AlN bulk for the L polarization and the wavevector q along the [001] high-symmetry direction is given by

$$\begin{aligned} D_{11} &= M_c^{-1}\Big[2(k_1+k_3)+2k_2\Big(1-\cos(qa/2)\Big)\Big] \\ D_{22} &= M_a^{-1}\Big[2(k_1+k_3)+2k_2\Big(1-\cos(qa/2)\Big)\Big] \\ D_{12} &= D_{21}^{*} = (M_aM_c)^{-1/2}\Big[2k_1\cos(qa/4)+2k_3\cos(3qa/4)\Big] \end{aligned} \tag{1}$$

where M_c and M_a denote the mass of cation and anion atoms, respectively, and a is the lattice constant. Diagonalization of the dynamical matrix D provides the eigenvalues and eigenvectors of the corresponding acoustic and optical vibrational modes.

Table 1. Fitted planar-force constants up to the third nearest-neighbours for cubic GaN and AlN.

	GaN (Nm^{-1})	AlN (Nm^{-1})
k_1	−17.577	−22.955
k_2	1.188	1.369
k_3	−0.042	−0.052

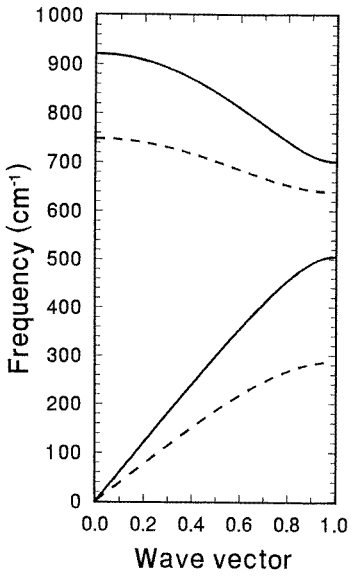

Figure 1. Calculated phonon dispersion curves of cubic GaN (solid lines) and AlN (dashed lines) in the L polarization for phonons propagating along the [001] direction.

In [2], phonons in cubic GaN and AlN were studied by a three-parameter empirical model: two accounting for the Keating-type potential and one for the effective charge responsible for the long-range Coulomb interactions. Due to the fact that there are no experimental data available up to now for cubic GaN and AlN, a folding procedure along

the [111] direction was used to derive the data from the experimental data of wurtzite counterparts. This is due to the similarity of the zinc-blende and wurtzite structures: the difference in lattice structures begins from the third neighbours. In the present work, for simplicity, a planar-force-constant model is used for L polarization along the [001] direction instead of three-dimensional calculations. The planar-force constants are obtained by fitting them to the phonon dispersion curves along the [001] direction in the L polarization given in [2]. It is found that the planar-force-constants up to the third nearest neighbours can produce satisfactorily the results obtained by the three-dimensional calculations in the framework of Keating potential together with the Coulomb interactions. The fitted planar-force constants for cubic GaN and AlN are given in table 1 and the resulting phonon dispersion curves in the L polarization for phonons propagating along the [001] direction are shown in figure 1.

For GaN/AlN SLs, the force constants across the interface are assumed to take on the average values of the two constituents. The dynamical matrix of the SL can be constructed with the given force constants. The dynamical matrix for SLs is given by

$$D(l,l') = (M_l M_{l'})^{-1/2} \sum_{l'} k_{nN+l'-l} \exp\left[\mathrm{i}qa(nN+l'-l)/4\right] \tag{2}$$

where l denotes the lth atomic layer in the unit cell of SL and $k_{nN+l'-l}$ is the planar-force constant between the l'th and the lth atomic layers; N is the periodicity of an SL and n is an integer. Because only the force constants up to the third nearest planar neighbour are considered, the following relation must be satisfied

$$| nN + l' - l | \leqslant 3. \tag{3}$$

The on-site interaction force constant ($l' = l$) can be obtained from the sum rule [3]

$$\sum_{l'} k_{nN+l'-l} = 0. \tag{4}$$

Solving the secular equation about the dynamical matrix, eigenvalues and eigenvectors for the SLs can be obtained.

In order to calculate the Raman intensity, a bond-polarizability model [4] is used. It has been found that this model can give a good description of the scattering intensity from optical modes. The polarizability of the whole system is calculated as a sum of the independent contributions from each bond, based on the calculated eigenvalues and eigenvectors. Then, the Raman intensity in the $\mu\nu$ polarization for the backscattering configuration is given by

$$I_{\mu\nu}(\omega) \propto [n(\omega)+1] \sum_j \delta(\omega - \omega_j(\boldsymbol{q}) |\Delta\alpha_{\mu\nu}(j\boldsymbol{q})|^2 \tag{5}$$

where $[n(\omega)+1]$ is the Bose–Einstein population factor.

The variation of the polarizability tensor $\Delta\alpha$ due to a phonon mode j with wavevector $\boldsymbol{q} \sim 0$ is a sum of the contributions from each bond [4]

$$\Delta\alpha = \begin{pmatrix} A_{xx} & A_{xy} & 0 \\ A_{xy} & A_{xx} & 0 \\ 0 & 0 & A_{zz} \end{pmatrix}. \tag{6}$$

For $(GaN)_m/(AlN)_n$ SLs, $A_{xx,zz}$ and A_{xy} are given by

$$A_{xx,zz} = 2(\alpha^{GaN}_{xx,zz} - \alpha^{AlN}_{xx,zz})(u_1 - v_1) \tag{7}$$

$$A_{xy} = (\alpha^{GaN}_{xy} + \alpha^{AlN}_{xy})(u_1+v_1) + 2\alpha^{GaN}_{xy} \sum_{i=2}^{2m} (-1)^{i-1} u_i + 2\alpha^{AlN}_{xy} \sum_{i=2}^{2n} (-1)^{i-1} v_i \tag{8}$$

where α is a fitted parameter and u_i and v_i are the displacements of ith atomic planes in GaN and AlN layers, respectively. For simplicity, the parameters α for GaN and AlN are assumed to be identical.

From (5) the well-known Raman selection rules can be built. Only the LO modes are Raman-active in the usual backscattering configuration. In the $z(x, y)\bar{z}$ scattering configuration, the LO modes with A_1 symmetry are active, while the modes with B_2 symmetry are active in the $z(x, x)\bar{z}$ scattering configuration. It can be seen from (7) and (8) that the Raman intensity for B_2 modes is usually small in the non-resonant condition.

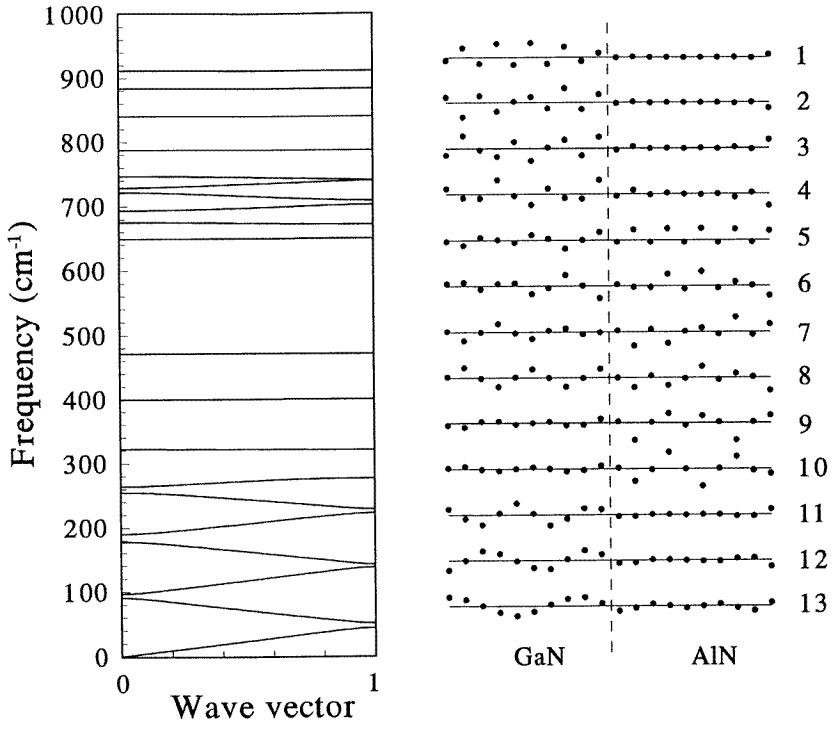

Figure 2. Calculated phonon dispersion curves of a (001)-oriented $(GaN)_5/(AlN)_5$ SL for phonons propagating along the [001] direction. The corresponding displacement patterns at the zone centre are also given. The modes are numbered from the top in order of decreasing frequency.

3. Calculated results

With the planar-force-constant model discussed above, the phonon dispersions of (001)-oriented $(GaN)_m/(AlN)_n$ SLs can be calculated, where m and n are the number of bilayers of GaN and AlN, respectively. To show the general features of phonons in SLs, the phonon dispersion curves of a $(GaN)_5/(AlN)_5$ SL in the L polarization are shown in figure 2 and the corresponding displacement patterns are also given. From figure 2 it can be seen that modes 1–4 are AlN-like LO confined modes. The vibrations are sharply confined to AlN layers. As a result, they are nearly dispersionless. It is interesting to note mode 5: strong excitations in GaN layers exist owing to the fact that its frequency overlaps the LO spectra continuum

of bulk GaN. It is resonant, quasi-confined to AlN layers. Modes 6–8 are resonant, quasi-confined GaN-like LO modes. Their frequencies are located in the LO continuum of bulk AlN. Strong excitations exist in AlN layers. As a result, there exist finite dispersions for these modes. Modes 9 and 10, with their frequencies outside the LO continuum of bulk AlN, are truly confined modes. The excitations in AlN layers are rather small. Modes 11–13 are also AlN-like confined modes, but they are from the bulk LA branches of AlN. The remaining modes are folded modes, from the LA branches of both constituents by a folding procedure. The small gaps at the zone-centre and zone-boundary are due to the difference in elastic constants of the two constituents.

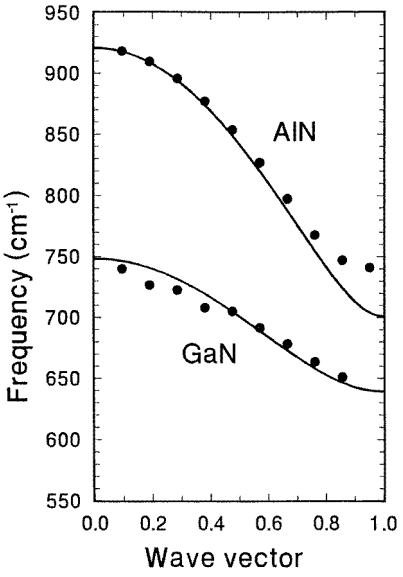

Figure 3. Comparison of the bulk phonon dispersions and the confined-mode frequencies of a $(GaN)_{10}/(AlN)_{10}$ SL mapped according to (9).

From the above discussions, it is seen that for the truly confined modes the vibrations are sharply confined to one of the constituent layers. As a consequence, the confined modes can be regarded as standing waves, described by an effective bulk wavevector [4, 5]

$$q_j = j\frac{\pi}{(n+\delta)d_0} \tag{9}$$

where n is the bilayer number, δ is a parameter describing the penetration of the vibrations of a confined mode into the adjacent material, $d_0 = a/2$ is the bilayer thickness, and j is the order of a confined mode. The atomic displacement of the jth mode along the layer normally has $j+1$ modes. The odd-order modes have A_1 symmetry, while the even order modes have B_2 symmetry. It should be indicated that a value of $\delta = 0.5$ corresponds to one monolayer. For a perfectly confined mode it is expected that $\delta = 0$. The establishment of an exact correspondence between the SL confined mode frequency and the bulk frequency is of use in two ways: firstly, it allows an accurate determination of the layer thickness; secondly, it could be used to determine the bulk phonon dispersion from the measurements on the SL modes as a function of j or n if δ is known. This idea has been used to determine the bulk phonon dispersions of those materials whose bulk dispersions are not known, like AlAs [6].

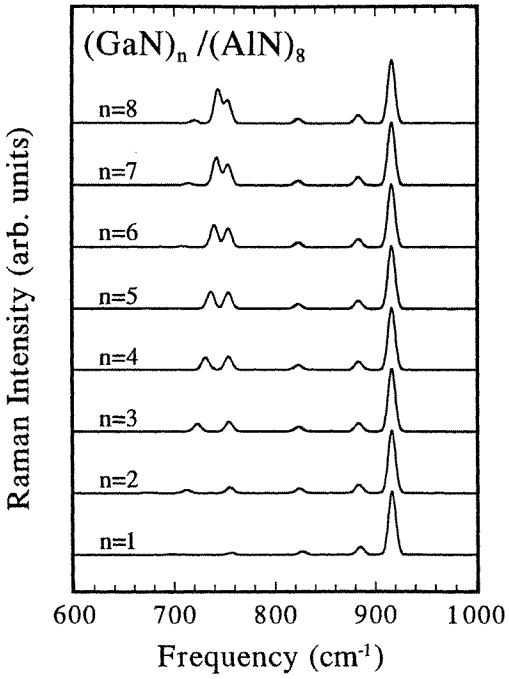

Figure 4. Calculated Raman spectra of (001)-oriented $(GaN)_8/(AlN)_n$ $(n = 1 - 8)$ SLs in the $z(x, y)\bar{z}$ scattering configuration.

This is especially meaningful for GaN and AlN since their bulk phonon dispersions are unknown.

It is found that for the truly confined modes a good correspondence can be established between the confined modes and the bulk dispersions provided that δ is properly chosen. We found that for both GaN and AlN, $\delta = 0.5$ can given a good correspondence between the bulk dispersions and the confined modes (see figure 3). This means that the vibrations of GaN or AlN confined modes extend one monolayer into the another constituent. From the figure it is obvious that there are some discrepancies in the correspondence for the AlN modes near the zone boundary and for the GaN modes near the zone centre. This is due to the fact that these modes are resonant, quasi-confined modes. Strong excitations exist in the adjacent constituent layers. In principle, they cannot be simply described by standing waves.

With the formulations given in the above section, the Raman spectra of GaN/AlN SLs can be calculated. The calculated Raman spectra of $(GaN)_n/(AlN)_8$ $(n = 1 - 8)$ SLs in the $z(x, y)\bar{z}$ configuration are given in figure 4. The four peaks above 740 cm^{-1} are from the AlN-like LO confined modes with odd order. From the Raman selection rules only the odd-order confined modes are active in the $z(x, y)\bar{z}$ configuration. The first-order mode (denoted by LO_1) has dominant Raman intensity. The intensities of the higher order modes are usually small. It is interesting to note the peak just below 750 cm^{-1} arising from the seventh-order AlN confined mode. With the increase of GaN layers the intensity of this mode is enhanced due to the strong excitations in GaN layers. For the GaN bilayers

increasing from 1 to 8, the AlN Raman peaks are nearly unchanged except for the peak just below 750 cm^{-1} . The appearance of the high-order confined modes is the symbol of confinement. It is interesting to note that only one GaN bilayer can cause a strong confinement of the vibrations of AlN. With the increase of GaN layers, the LO_1 peak of GaN increases in intensity. Its peak position shifts upwards a little. This can be understood by the fact that the bulk LO branch has a small dispersion (see figure 1).

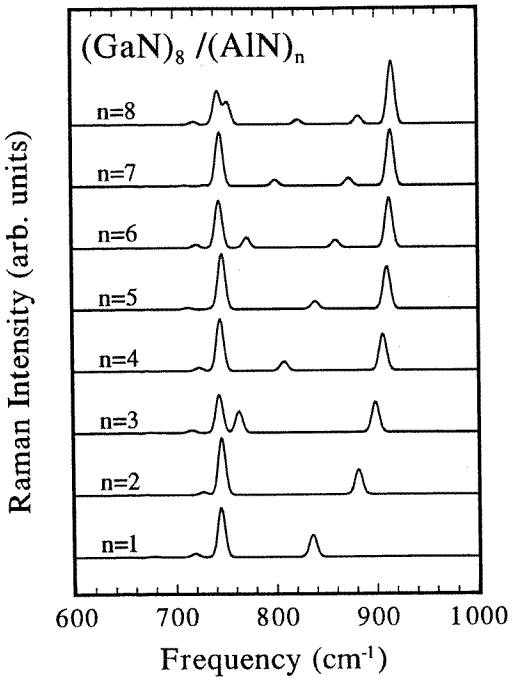

Figure 5. Calculated Raman spectra of (001)-oriented $(GaN)_n/(AlN)_8$ $(n = 1 - 8)$ SLs in the $z(x, y)y\bar{z}$ scattering configuration.

In figure 5 the calculated Raman spectra of $(GaN)_8/(AlN)_n$ $(n = 1 - 8)$ SLs are shown. With the increase of AlN layers, the LO_1 peak shifts upwards in frequency. When $n \geqslant 3$, the LO_3 peak appears, and the LO_5 peak is present when $n \geqslant 5$. The intensities of the high-order GaN-like LO modes are very small, and it is difficult to get the confinement from the appearance of the high-order confined modes. This can, however, be obtained by an analysis of the displacement patterns. It is found that one bilayer of AlN can also cause strong confinement for the vibrations of GaN layers.

4. Conclusions

The phonon properties of (001)-oriented cubic GaN/AlN in the L polarization were studied by a planar-force-constant model. Truly confined and resonant, quasi-confined AlN-like LO modes were found. GaN-like LO modes are quasi-confined or confined depending on whether their frequencies overlap with the bulk LO continuum of AlN or not. For both GaN and AlN confined modes their frequencies can be obtained simply from the bulk dispersion

with the effective wavevector $q_j = j\pi/[(n + 0.5)d_0]$, where j is the order of the confined mode, and n and d_0 are the number of bilayers and bilayer spacing, respectively. Raman spectra were calculated by the bond-polarizability model. It was found that one bilayer of GaN (AlN) can cause a strong confinement of the vibrations of AlN (GaN) layers.

Acknowledgments

Support from the Qi-Ming Research Plan of Shanghai and NSF of China is acknowledged.

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