Lattice Dynamics of GaSb/AlSb Strained Superlattices

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Abstract – Phonon modes of short-period GaSb/AlSb superlattices are calculated from first principles within density functional perturbation theory. Strain effects in the superlattices layers are taken into account directly by preliminary optimization of their structural parameters. Calculated phonon frequencies are compared with experimental ones, obtained from Raman and infrared spectroscopies. It is demonstrated that strain in superlattices layers, originated from bulk materials lattice constants mismatch, gives rise to additional shift of 1–3 cm^{-1} in observable in Raman and infrared spectra frequencies in comparison with frequencies of the bulk components.

A study of vibrational states of semiconductor superlattices (SLs) is an actual problem in connection with their important role in formation of optical and electrical properties of materials, and due to their high sensitivity to structural defects.

SLs GaSb/AlSb attract the steadfast attention of researchers in connection with their possible applications to optoelectronic devices, for example in laser diodes and injection lasers fabrication. These systems were studied mainly experimentally, for example, in [1–4] and to a lesser extent theoretically [5–6]. In the present paper systematic first principles calculations of phonon modes in short-period $(GaSb)_n(AlSb)_n$ (001) SLs with number of monolayers $n = 1–10$ are carried out. The obtained results are in good agreement with experimental data.

Phonon calculations were performed within density functional perturbation theory (DFPT) which detailed review can be found in [7], with use of nonlocal norm-conserving pseudopotentials [8] and plane wave basis set. Exchange-correlation effects were treated in the local density approximation. All numerical results, described in the present paper, were obtained by means of software package *Quantum Espresso* [9].

Electronic wave functions were expanded in plane waves with energy up to a cutoff of 20 Ry. Brillouin zone integration was performed with using of the Monkhorst–Pack method of special points [10] on a 6×6×6 mesh for bulk crystals and monolayer SL and on meshes 6×6×2 for $(GaSb)_2(AlSb)_2$, $(GaSb)_3(AlSb)_3$ and 6×6×1 for other studied SLs. Decreasing of the *k*-points number along a *Z*-axis reflects a reduction of the Brillouin zone when the number of layer in the SL increases. The theoretical values of GaSb and AlSb lattice constants, derived at given parameters, corresponding to a minimum of the crystal total energy, are 6.067 and 6.109 Å, that less than 0.5% differs from the experimental values ($a_{GaSb} = 6.096$ Å, $a_{AlSb} = 6.136$ Å [11]). The difference between theoretical values of lattice constants (0.69%) is rather close to the difference of their experimental values (0.65%) that indicates high quality of the generated pseudopotentials for the gallium and the aluminum, correctly describing properties of these chemical elements in GaSb and AlSb.

The difference in the values of bulk composites lattice constants leads to a strain occurrence in SL layers. So, GaSb layers are stretched, and AlSb layers are squeezed. It results to a lower symmetry (C_{2v}), in comparison with symmetry of lattice matched GaAs/AlAs or GaP/AlP SLs (D_{2d}). Therefore, before performing vibrational modes calculations, preliminary optimization of SLs structural parameters was carried out. The values of lattice constant *a*, obtained as a result of structural optimization, for different *n* practically coincide with an arithmetic average of a_{GaSb} and a_{AlSb}.

Calculations were performed for SLs $(GaSb)_n(AlSb)_m$ with identical number of monolayers $n = m$ of different components, therefore this coincidence is the consequence of proximity of GaSb and AlSb elastic constants.

First principles calculations of the phonon dispersion curves of bulk crystals GaSb, AlSb and another binary semiconductors have been reported in [12] and show close agreement to the experimental data, obtained by a method of neutron spectroscopy. Using the same implementation of the linear response method, we have obtained analogous results. Comparison with experiment also shows high quality of the calculation of bulk components phonon spectra. Thus, for GaSb the frequencies of transverse and longitudinal optical phonons (TO and LO) computed by us in the Brillouin zone centre $\omega_{TO} = 232$ cm^{-1}, $\omega_{LO} = 240$ cm^{-1}, whereas in according with presented in [2] experimental data $\omega_{TO} = 230$ cm^{-1}, $\omega_{LO} = 239$ cm^{-1}. For AlSb our calculations give values $\omega_{TO} = 324$ cm^{-1}, $\omega_{LO} = 341$ cm^{-1}, and in experiment frequencies $\omega_{TO} = 323$ cm^{-1}, $\omega_{LO} = 344$ cm^{-1} [2] are determined.

The vibration spectrum of monolayer $(GaSb)_1(AlSb)_1$ SL, reflecting all main peculiarities in spectra of the SLs with various number of layers, is depicted in Fig. 1 along symmetrical directions. In Table 1, the calculated frequencies are compared with the measured ones [4].

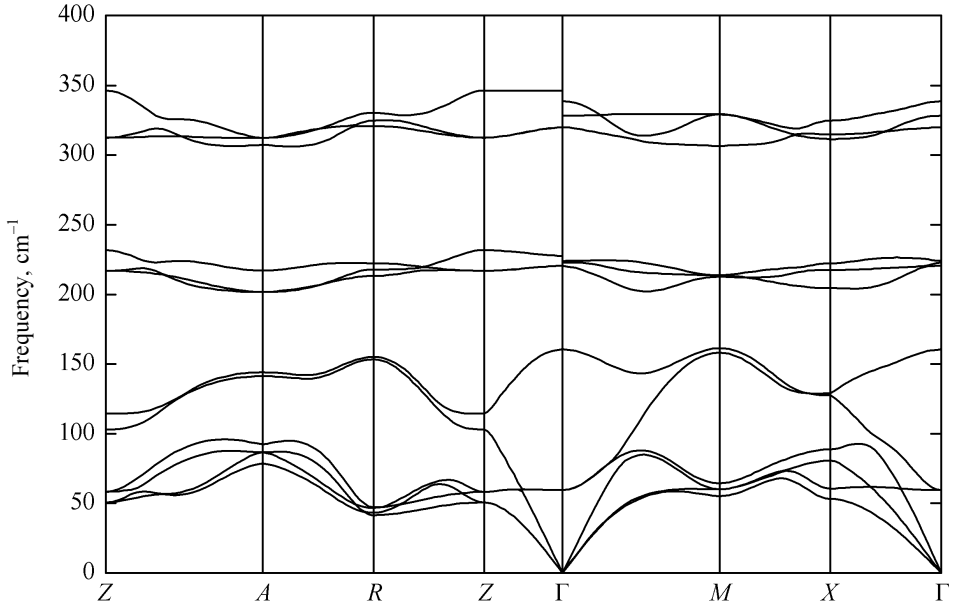

Fig. 1. Phonon dispersion curves of $(GaSb)_1(AlSb)_1$ SL

Table 1. Frequencies of GaSb- and AlSb-like TO and LO modes in $(GaSb)_n(AlSb)_n$ SLs, cm^{-1}

Monolayers number	GaSb-like modes				AlSb-like modes			
	Theory		Experiment IR (Raman) [4]		Theory		Experiment IR (Raman) [4]	
n	TO	LO	TO	LO	TO	LO	TO	LO
1	220	228			320	346		
2	224	236	220	230 (235)	320	345	317	337 (338)
3	226	237			323	344		
4	228	238	223 (226)	234 (234)	324	344	319	339 (343)
5	228	238			324	344		
7	229	238	225 (227)	236 (237)	325	344	319	340 (342)

In Fig. 2, phonon densities of states of $(GaSb)_1(AlSb)_1$ SL and bulk materials are compared. As is seen from this figure, the density of states of monolayer SL mainly keeps the basic features of densities of states of GaSb and AlSb.

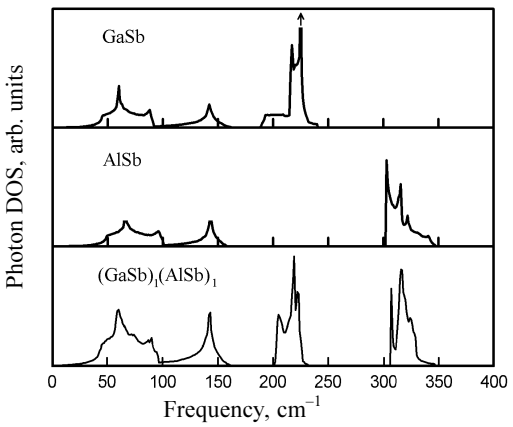

Fig. 2. Phonon density of states of bulk crystals GaSb, AlSb, and $(GaSb)_1(AlSb)_1$ SL

The tetragonal cell of monolayer SL contains four atoms, therefore a phonon spectrum of $(GaSb)_1(AlSb)_1$ consists of 12 separate branches. Apparently, from Fig. 1, in a spectrum of the monolayer SL it is possible to separate three intervals of the allowed frequencies, the upper bound is at 346 cm^{-1}. Acoustic vibrations of bulk crystals GaSb, AlSb are in an equal interval of the frequencies, therefore a new periodicity imposed by sequence of layers of different bulk components in the SL, does not influence on acoustic phonons propagation. The origin of vibrational modes of the low frequency range (to 160 cm^{-1}) is related to folding of acoustic branches of bulk components owing to decreasing of SL's Brillouin zone.

Two phonon bands at intervals of 200–250 and 300–350 cm^{-1} consisting of three branches, are caused by the vibrations localized in SL layers. As the analysis of polarization vectors shows, the upper band of optical phonons conforms to AlSb vibrations, lower one to GaSb vibrations. The gap between them is related to difference of cation masses.

Figure 3 shows the dependencies of $(GaSb)_n(AlSb)_n$ SLs TO and LO phonon frequencies on the number of monolayers n. The line connecting theoretical values is drawn for convenience. Experimental values are marked by quadrates and circles: the solid symbols are data obtained by Raman and open ones are by infrared spectroscopies [4]. As one can see from this figure and Table 1, our values are closer to the results of Raman scattering measurements [4].

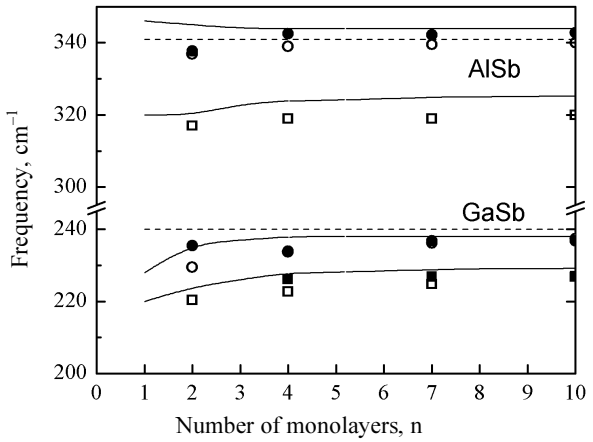

Fig. 3. Dependencies of $(GaSb)_n(AlSb)_n$ SLs TO and LO phonon frequencies on the number of monolayers n. The solid lines connect the theoretical points. Quadrates and circles are experimental data from [4]

Influence of confinement effects on a phonon spectra lattice matched SLs GaAs/AlAs have been well studied and manifested in decreasing of GaAs- and AlAs-like phonon modes frequencies in comparison with their values in bulk crystals. If lattice constants of bulk components do not match, which is the case for systems under study, additional shift of phonon frequencies occurs due to a strain in SL layers. Influence of these effects is different: for GaSb-like modes, due to a stretching of GaSb layers in SL, both of these effects are summarized, that leads to decreasing of the frequencies, whereas in case of AlSb-like modes their action is opposite since AlSb layers are shrinking in GaSb/AlSb SLs. This mechanism determines a view of the curves plotted in Fig. 3. One can see from Fig. 3 that weakening of confinement effects with increasing the number of stacked monolayers does not lead to the achieving of LO phonon frequencies of bulk composites (dashed line) that is the consequence of the influence of a strain remaining. It should be mentioned, that actually already at $n = 4$ and $n = 7$ there is a stabilization of LO and TO modes frequencies, respectively. Thus, the shift from bulk components frequencies is fixed at level of 1–3 cm^{-1}.

Our results have systematically higher values, compared with available experimental data. The origin is imperfection of real heterostructures. Interface layers of SLs due to intermixing of atoms of metals are closer in structure to solid solutions with different state of disorder. A degree of interface layers ordering is a very important quality index for grown heterostructures. In the presence of interface roughness, the higher-indexed modes are substantially affected. Decreasing of an effective thickness of structural ordering layers leads to decreasing of obtained experimentally localized modes frequencies in comparison with their frequencies in ideal SL. The transmission electron microscopy experiments performed in [4], showed intermixing of atoms at the interface region in the $(GaSb)_4(AlSb)_4$ SL of 1–2 monolayers. As our calculations shows this structure imperfection is quite sufficient for influence on the phonon frequencies.

Thus, the present calculations from first principles allowed us to perform numerical modeling of phonon spectra of semiconductor GaSb/AlSb SLs. Both confinement and strain effects in the SLs layers have been naturally taken into account. The obtained results can be used in the analysis of experimental data for studied heterostructures, for example, at estimating the interface properties.

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