Phonons and periodons in IV–VI semiconductor superlattices

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Abstract. We have calculated the phonon and periodon dispersion relations in IV–VI semi-conducting bulk PbTe and SnTe and their superlattice structure. The model used here is a one-dimensional lattice which includes harmonic interactions up to second neighbours as well as on-site nonlinear electron-ion interactions at the anion site. We calculate the phonon and periodon dispersion relations in bulk and PbTe–SnTe superlattice for the transverse optic and acoustic modes using the transfer matrix method. Our analysis has predicted correct nature of the folding of acoustic and confinement of optical phonons at various frequency intervals corresponding to pass and stop bands of the superlattices.

1. Introduction

During the last few decades, the ferroelectric IV–VI compounds (PbTe, SnTe) have generated a considerable interest in structural phase transition and mode softening [1–5]. These compounds exhibit soft mode behaviour at the zone centre and zero gap transition, depending on temperature and composition of the alloy systems. The ferroelectric [4, 5] and phonon [6] properties of IV–VI compounds have been investigated extensively in the recent past. It is known from literature that the transverse optical branches of these compounds show some anomalous behaviour in the middle of the $[0, 0, \xi]$ direction. This occurs because of the presence of free carriers due to intrinsic defects in the crystals. Bilz *et al* [7] have calculated the transverse phonon modes by using a simple diatomic linear chain model with first and second nearest neighbour core-core interactions and nonlinear polarizability at the chalcogen ion site. They have also extended their treatment to obtain an exact solution which can describe the phonon anomalies, electro-optic phenomena and the nonlinear excitations such as periodons [7]. In order to understand the interactions between the phonons and periodons, Bussmann-Holder have used an isotropic monoatomic polarizable linear chain model and calculated the periodon amplitude and dispersion curves [7].

It is revealed from literature that apart from the investigation of the electronic and dynamical properties of the bulk materials, modulated structures (i.e., superlattices) of these compounds are also investigated [8–10]. In the present paper we report the calculated vibrational properties of the less investigated IV–VI semiconducting super-lattices (PbTe–SnTe). For this system, only a comparative study of the structural and superconducting properties of epitaxial grown superlattices (PbTe–SnTe, PbTe–SnS)

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has been reported [9] and also galvanomagnetic properties are studied [10]. In the superlattice structure, it is expected that the IV–VI compounds will manifest a variety of nonlinear phenomena because of the non-linear polarizability of chalcogen ion.

In the present paper we have calculated the phonon and periodon dispersion relation in bulk PbTe and SnTe and their superlattice structure. We have developed a nonlinear lattice dynamical theory which includes the effect of first neighbour on-site and inter-site and second neighbour inter-site interactions in the one-dimensional diatomic chain model with nonlinear fourth order polarization potential at the anion site. Using this model, the phonon and periodon dispersion curves for transverse acoustic and optic modes are calculated without using self consistent phonon approximation. We have used the transfer matrix approach for calculating the dispersion curves in PbTe–SnTe superlattices, both for phonons and periodons.

2. Theory

2.1 *Bulk semiconductors*

In bulk structure, our model consists of a one-dimensional diatomic chain in which the cations (Pb, Sn) are assumed to be linearly polarizable and the anions (Te) are
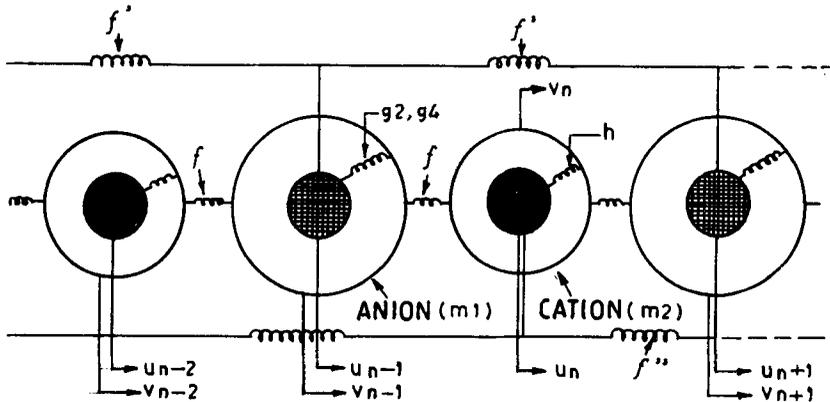

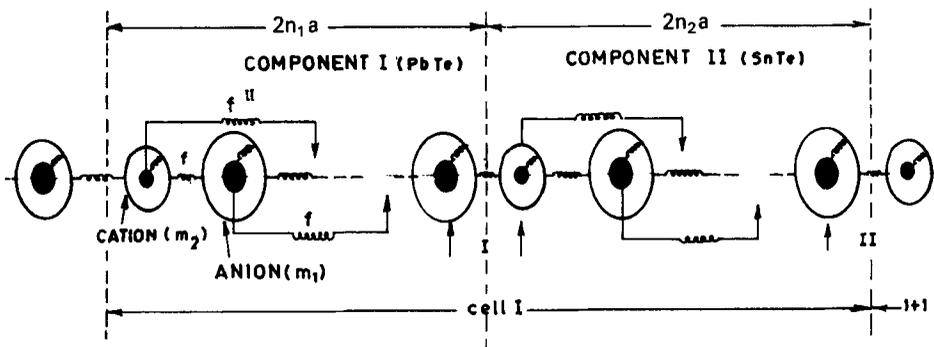**Figure 1(a).** One-dimensional ϕ^4 linear chain model with nearest and next nearest neighbour interactions.

Figure 1(b). Linear chain model of a one-dimensional superlattice with nearest and next nearest neighbour interactions.

assumed to be nonlinearly polarizable, which are configurationally unstable in nature. Anionic shell-core coupling consists of an attractive harmonic (g_2) and a fourth order repulsive force constants (g_4). The one-dimensional diatomic lattice with different types of interactions is shown in figure 1(a). f is the first neighbour interaction between the shells of anion and cation and f' and f'' are the force constants due to second neighbour interactions between the cores of cation and anion. h is the attractive cationic force constant.

In view of this, the equation of motion can be written as

$$m_1 \ddot{u}_{n-1} = g_2 w_{n-1} + g_4 w_{n-1}^3 + f'(u_{n+1} + u_{n-3} - 2u_{n-1}) \tag{1}$$

$$m_2 \ddot{u}_n = f''(u_{n+2} + u_{n-2} - 2u_n) + h w_n \tag{2}$$

$$m_{e_1} \ddot{v}_{n-1} = -g_2 w_n - g_4 w_{n-1}^3 + f(w_n + w_{n-2} - 2w_{n-1}) +$$
$$f(u_n + u_{n-2} - 2u_{n-1}) \tag{3}$$

$$m_{e_2} \ddot{v}_n = -h w_n + f(w_{n+1} + w_{n-1} - 2w_n) + f(u_{n+1} + u_{n-1} - 2u_n) \tag{4}$$

where m_1, m_2 and $m_{e(i)}$ ($i = 1, 2$) are the masses of anion, cation and the valence electrons of anion and cation respectively. $u_{n\pm 1, n\pm 3, n\pm 5,...}$ are the displacements of the cores of anion and $u_{n,n\pm 2, n\pm 4,...}$ are the displacements of the cores of cation in the linear infinite chain. By considering adiabatic condition (i.e., $m_e \approx 0$), the LHS of (3) and (4) vanishes. Also we define a relative core-shell coordinate as $w_{n-1(n)} = v_{n-1(n)} - u_{n-1(n)}$.

For calculating the phonon dispersion, both the anions and cations are considered to be linearly polarizable (i.e., $g_4 = 0$). We assume the following solutions for solving the equations of motion. For the anions, the solutions are

$$w_{n-1} = A \sin(\omega t - (n-1)qa), \tag{5}$$

$$u_{n-1} = B \sin(\omega t - (n-1)qa). \tag{6}$$

For cation we write the displacements as

$$u_n = \alpha u_{n-1} \tag{7}$$

where α is an arbitrary constant. Equations (3) and (4), together with above considerations results to

$$w_{n-1} = \frac{f}{(g_2 + 2f)}[(u_n + u_{n-2} - 2u_{n-1}) + (w_n + w_{n-2})], \tag{8}$$

$$w_n = \frac{f}{(h + 2f)}[(u_{n+1} + u_{n-1} - 2u_n) + (w_{n+1} + w_{n-1})]. \tag{9}$$

Substituting (5), (6), (7) in (8) one gets the ratio of the amplitudes as

$$\frac{A}{B} = \frac{2f}{(g_2 + 4f \sin^2(qa/2))}(\alpha \cos qa - 1). \tag{10}$$

Equation (9) also results in the following amplitude ratio

$$\frac{A}{B} = \frac{2f}{(h + 4f \sin^2(qa/2))}(\cos qa - \alpha). \tag{11}$$

The constant α which relates the displacements of the atoms of the neighbouring unit cells can be obtained by substituting (5), (6), (7) and (10) in (7) as

$$\alpha = \frac{\left[-m_1\omega^2 + 4f'\sin^2(qa/2) + \dfrac{2g_2 f}{(g_2 + 4f\sin^2(qa/2))} \right]}{\dfrac{2g_2 f \cos qa}{(g_2 + 4f\sin^2(qa/2))}} \tag{12}$$

Similarly from (2) α can be obtained by substituting (5), (6), (7) and (11) in (7) as

$$\alpha = \frac{\dfrac{2hf\cos qa}{(h + 4f\sin^2(qa/2))}}{\left[-m_2\omega^2 + 4f''\sin^2(qa/2) + \dfrac{2hf}{(h + 4f\sin^2(qa/2))} \right]}. \tag{13}$$

Equating (12) and (13) we obtain the phonon dispersion relation for bulk as

$$\omega^4 + Z\omega^2 + Y = 0 \tag{14}$$

with

$$Z = -\left[\frac{1}{m_1}\left(4f'\sin^2(qa/2) + \frac{2g_2 f}{(g_2 + 4f\sin^2(qa/2))} \right) + \right.$$
$$\left. \frac{1}{m_2}\left(4f''\sin^2(qa/2) + \frac{2hf}{(h + 4f\sin^2(qa/2))} \right) \right] \tag{15}$$

$$Y = \frac{1}{m_1 m_2}\left[\left\{ \left(4f'\sin^2(qa/2) + \frac{2g_2 f}{(g_2 + 4f\sin^2(qa/2))} \right) \cdot \right. \right.$$
$$\left. \left(4f''\sin^2(qa/2) + \frac{2hf}{(h + 4f\sin^2(qa/2))} \right) \right\} -$$
$$\left. \frac{4g_2 hf^2 \cos^2 qa}{(g_2 + 4f\sin^2(qa/2))(h + 4f\sin^2(qa/2))} \right]. \tag{16}$$

To obtain the periodon dispersion relation, we have considered the anions to be nonlinearly polarizable and cations to be linearly polarizable and used adiabatic approximation. Bilz [7] has already shown that the periodon frequencies are sub-harmonics of the corresponding phonon eigen frequencies with equal scaling by a factor of three which reduces the Brillouin zone into one third of its value. Exact nonlinear periodic solutions can be expressed as

$$w_{n-1} = A\sin(\omega t - (n-1)qa) \tag{17}$$

$$u_{n-1} = B\sin(\omega t - (n-1)qa) + C\sin 3(\omega t - (n-1)qa). \tag{18}$$

The ratio of the amplitudes can be derived from (1) and by equating the coefficients of $\sin(\omega t - (n-1)qa)$ as

$$\frac{A}{B} = \frac{-m_1\omega^2 + 4f'\sin^2(qa/2) - 2f(\alpha\cos qa - 1)}{-4f\sin^2(qa/2)}. \tag{19}$$

Equating the coefficients of $\sin 3(\omega t - (n-1)qa)$ in (1), the constant α is defined as

$$\alpha = \frac{-9m_1\omega_P^2 + 4f'\sin^2(3qa/2) + 2f}{2f\cos 3qa} \tag{20}$$

where ω_P is the periodon frequency.

Similar sets of equation can also be obtained for the amplitude ratio A/B by using (2) for mass m_2. This can be done by equating the coefficients of $\sin(\omega t - (n-1)qa)$ and is given by

$$\frac{A}{B} = \frac{-m_2\alpha\omega^2 + 4f''\sin^2(qa/2) + \dfrac{2hf}{h+2f}(\cos qa - \alpha)}{\dfrac{2hf}{(h+2f)}\cos qa}. \tag{21}$$

For the periodon dispersion, the value of α obtained from (2) as

$$\alpha = \frac{-4f''\sin^2(3qa/2) + \dfrac{2hf}{(h+2f)}\cos 3qa}{-9m_2\omega_P^2 + \dfrac{2hf}{(h+2f)}}. \tag{22}$$

The dispersion relation for the periodon in the diatomic chain can be obtained by equating the values of α (from (20) and (22)) obtained for two different masses as

$$\omega_P^4 + Z'\omega_P^2 + Y' = 0 \tag{23}$$

with

$$Z' = -\frac{2}{9}\left[\frac{1}{m_1}(2f'\sin^2(3qa/2) + f) + \frac{1}{m_2}\left(\frac{hf}{(h+2f)}\right)\right] \tag{24}$$

$$Y' = \frac{4}{81m_1m_2}\left[(2f'\sin^2(3qa/2) + f)\left(\frac{hf}{(h+2f)}\right) - \right.$$
$$\left. f\cos 3qa\left(-2f''\sin^2(3qa/2) + \frac{hf}{(h+2f)}\cos 3qa\right)\right] \tag{25}$$

and the squared amplitude is obtained from (3), (17), (18) and (19) as

$$A^2 = \frac{4g_2}{3g_4}\left[1 + \frac{4f}{g_2}\left(\frac{m_1\omega_P^2 - 4f'\sin^2(qa/2) + f(\alpha\cos qa - 1)}{m_1\omega_P^2 - 4f'\sin^2(qa/2) + 2f(\alpha\cos qa - 1)}\right)\sin^2(qa/2)\right]. \tag{26}$$

2.2 *Superlattice structure*

The layered structure or superlattice consists of alternating layers of thickness d_1 of the constituent I and of thickness d_2 of constituent II. Different specimens can be prepared so that d_1 and d_2 have any value from two or three atomic spacing up to the order of 100 nm. For simplicity, we have assumed that the interatomic lattice spacing (a) and the elastic constants (c_{ij}) have the same values in both the components of the periodically layered structures. The physical properties of the layered structures

are greatly modified as compared to the bulk materials, due to the increase in the spatial period $(D = 2(n_1 + n_2)a)$. As a consequence a new Brillouin zone at the wavevector π/D is developed perpendicular to the interfaces. A schematic diagram for a one-dimensional diatomic bilayer superlattice with different force constants is shown in figure 1(b). The derivation of phonon dispersion relation in superlattices has been given in detail in [11] by using the transfer matrix method. The dispersion relation for bilayer compound semiconductor superlattice is given by [12]

$$\cos QD = \cos q_1 d_1 \cos q_2 d_2 +$$
$$\left[1 - \frac{1}{2}\left(\frac{\alpha_1}{\alpha_2} + \frac{\alpha_2}{\alpha_1}\right) \frac{1}{\cos q_1 a \cos q_2 a} \right] \frac{\sin q_1 d_1 \sin q_2 d_2}{\tan q_1 a \tan q_2 a}. \qquad (27)$$

For the calculation of phonon dispersion curves in semiconductor superlattice, $\alpha_i(i = 1, 2,$ for two different layers) are obtained from (12) and (13) and the phonon wave vectors q_1 and q_2 as a function of frequency are obtained from (14).

Similarly the periodon dispersion relation in superlattice structure, is obtained for the different periodon wave vectors q_1 and q_2 as a function of frequency from (23) and the values of α_i from (20) and (22). In the case of phonon dispersion, a new Brillouin zone appears with wavevector $QD = 2\pi$, which becomes twice the Brillouin zone in bulk (i.e., $qa = \pi$). As mentioned before, the frequency of the periodon becomes maximum at one third of the phonon wave vector. Therefore for the periodon dispersion, the boundary of the Brillouin zone lies at $2\pi/3$ value of the phonon wave vector.

3. Results and discussion

The polarizability model developed in the theory calculates the dispersion curves for the transverse acoustic and ferroelectric transverse optic mode. In the IV–VI semiconductor, we have calculated the phonon and periodon's transverse branches by fitting the model parameters f, f', f'', g_2, h and g_4 to the experimental zone centre frequencies. These branches are of quite interest as it undergoes mode softening at the ferroelectric phase transition of these compounds. In the present case, SnTe undergoes a transition to a ferroelectric state with a finite transition temperature of 98 K, whereas PbTe does not undergo phase transition and remain paraelectric for all temperatures. Also the soft mode concept is extended to the periodon case to understand the coupling between nonlinear modes and normal phonon modes.

In figures 2(a) and (b), we have plotted the transverse optic and acoustic branches of the phonon dispersion relations for bulk (PbTe and SnTe) (shown in figure 2(a)) and their superlattice structure (PbTe–SnTe), consisting each of six layers (shown in figure 2(b)). We have also shown in figure 2(a), the experimental points for bulk PbTe at 293 K taken from ref. [13] and SnTe at 100 K from ref. [4, p-90]. The corresponding parameters for the calculation of the phonon dispersion curves are defined in table 1. In the phonon dispersion curves of bulk PbTe and SnTe, we have observed some discrepancies in the calculated and experimental values of the transverse optic mode around the wavevector $(0, 0, 0.3)$ to $(0, 0, 0.8)$. This is mainly because our present analysis does not consider the effect of screening due to the free carriers in the systems. This fact however, is not a serious limitation of our predicted results, for such a simple model which can qualitatively explain the nature of the branches in the dispersion curves. In adjoining figure 2(b) we have shown the phonon dispersion curves (TO and TA) for $(PbTe)_6$–$(SnTe)_6$ superlattices.
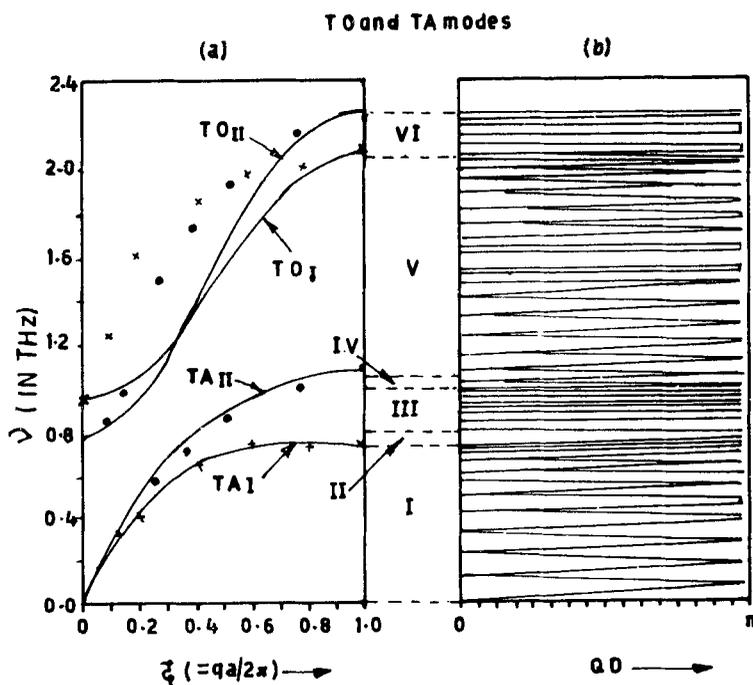
Figure 2(a). Bulk phonon dispersion curves of transverse acoustic and optic branches of PbTe (I) and SnTe (II). v $(=\omega/2\pi)$ is the reduced frequency and $\xi(=qa/2\pi)$ is the reduced wavevector. Experimental points for PbTe [×] and SnTe [●] are taken from ref. [13] and [4, p. 90] respectively.

Figure 2(b). Phonon dispersion curves of transverse acoustic and optic branches of PbTe–SnTe superlattice with $n_2 = n_1 = 6$.

Table 1. Parameters for phonon dispersion in bulk (PbTe and SnTe).

Parameters	Value	
	SnTe	PbTe
$f(10^4 \text{g/s}^2)$	0·1237	0·2385
$f'(10^4 \text{g/s}^2)$	0·997	0·745
$f''(10^4 \text{g/s}^2)$	0·1655	0·085
$g_2(10^4 \text{g/s}^2)$	− 59.9	− 55·9
$h(10^4 \text{g/s}^2)$	− 111·246	− 103·0
m_1(a.m.u.)	127·5	127·5
m_2(a.m.u.)	118·7	207·2

To interpret the results of bulk and superlattice dispersion curves, we have divided the frequency axis into six regions on the basis of the values of the real and complex wavevectors of the components I (PbTe) and II (SnTe), which is indicated in table 2. This type of frequency division was given in ref. [12, p-260] for simple superlattice structure. In region I, both the wavevectors q_{TA_I} and $q_{TA_{II}}$ are real, which implies that in very low frequency dispersion region, we obtain broad pass and narrow stop bands.

Table 2. Frequency intervals of figure 2(a).

Frequency interval	q_{TA_I}	$q_{TA_{II}}$	q_{TO_I}	$q_{TO_{II}}$	Nature of superlattice phonon dispersion curves.
I	R	R	I	I	Broad pass and narrow stop acoustic bands.
II	C	R	I	I	Broad stop and narrow pass acoustic bands.
III	I	R	I	R	Broad stop acoustic and optic modes.
IV	I	R	R	R	Broad pass bands for optic modes and broad stop bands for acoustic modes.
V	I	I	R	R	Narrow stop and broad pass bands for optic modes.
VI	I	I	I	R	Broad stop optic bands.

R = real, C = complex, I = pure imaginary
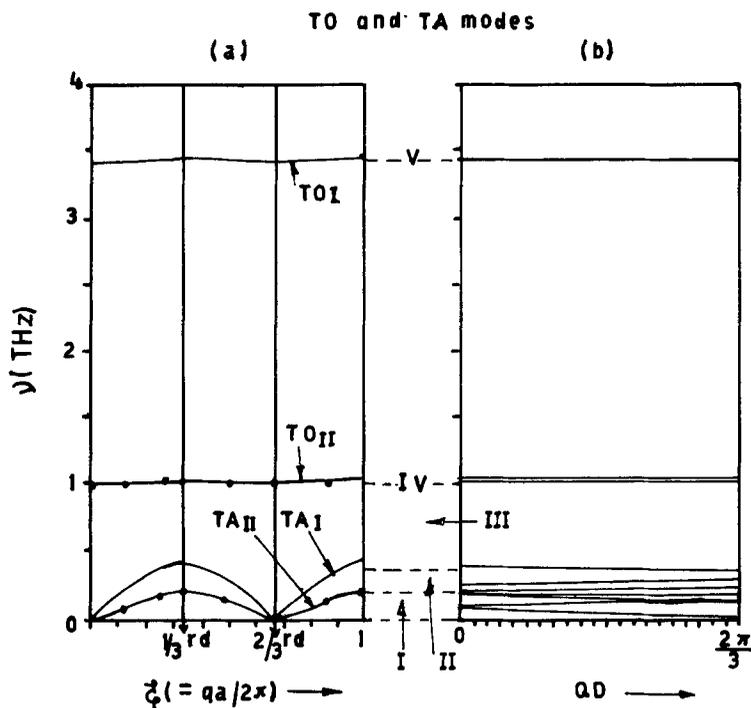

TO and TA modes

Figure 3(a). Bulk periodon dispersion curves of transverse acoustic and optic branches of PbTe (I) and SnTe (II). Model calculation results of SnTe [●] are taken from ref. [4] p. 90.

Figure 3(b). Periodon dispersion curves of transverse acoustic and optic branches of PbTe–SnTe superlattice with $n_2 = n_1 = 6$.

Hence the modes are propagating in nature in the frequency interval $0 < v < 0.6$ THz. From $v = 0.6$ to 0.75 THz., the wavevectors q_{TA_I} remains constant from 0.4 to 1.0 whereas the wavevector $q_{TA_{II}}$ increases from 0.25 to 0.475. In this frequency interval, the modes are propagating as well as confined in some of the regions. In region II (i.e., $0.75 < v < 0.78$), only $q_{TA_{II}}$ is real which results into broad stop and narrow pass bands. The transverse acoustic modes of SnTe become confined in this frequency interval. In region III (i.e., $0.78 < v < 0.96$) the wavevectors $q_{TO_{II}}$ and $q_{TA_{II}}$ are only real, which shows that we obtain broad stop bands in this region. In region IV (i.e., $0.96 < v < 1.05$) three wavevectors i.e., $q_{TA_{II}}$, q_{TO_I} and $q_{TO_{II}}$ are real, which signifies that the optical modes are propagating in nature and the acoustic modes of SnTe remain confined. In region V, the optical modes in both the materials are real. The frequency varies from 1.05 to 2.075 and the dispersion obtained there consists of narrow stop and broad pass bands. In the region VI, only the wavevector $q_{TO_{II}}$ is real in magnitude. As expected we obtained there broad stop bands.

In figures 3(a) and (b), we have obtained the periodon dispersion relation in bulk and superlattice structure of PbTe and SnTe layers. From the figure 3(a), it is noted that the transverse optic modes of periodon remain constant for all the wavevectors and the transverse acoustic modes become maximum at one third of the reduced wavevector [4]. Hence in the periodon treatment, the Brillouin zone reduces to one third of the phonon case. For the material SnTe, Bilz *et al* [7] have calculated the periodon dispersion relation in which it is found that the LO phonon become degenerate with the TO phonon at the zone centre. The periodon frequency obtained from their calculation are also shown in figure 3(a) for SnTe material. For PbTe, no such measurements are available. Corresponding to the wavevector obtained in bulk,

Table 3. Parameters for periodon dispersion in bulk.

Parameters (10^4 g/s^2)	Value	
	SnTe	PbTe
f	3.8145	44.302
f'	0.012	0.05
f''	0.073	0.53
h	-0.0777	-0.9

Table 4. Frequency intervals of figure 3(a).

Frequency interval	q_{TA_I}	$q_{TA_{II}}$	q_{TO_I}	$q_{TO_{II}}$	Properties of superlattice periodon dispersion curves.
I	R	R	I	I	Broad pass and narrow stop acoustic bands.
II	C	R	I	I	Broad stop bands for acoustic modes.
III	I	I	I	I	No solutions.
IV	I	I	C	R	Broad stop optic bands.
V	I	I	R	C	Broad stop optic bands.

R = real, C = complex, I = Imaginary

we have obtained the periodon dispersion relation which is shown in figure 3(b). The corresponding model parameters and the frequency interval are given in tables 3 and 4 respectively. In region I (i.e., from $0 < v < 0.1875$), the wavevectors q_{TA_I} and $q_{TA_{II}}$ are real which give rise to propagating nature of the acoustic modes. In region II (i.e., from $0.1875 < v < 0.375$) only the wavevectors $q_{TA_{II}}$ is real. The acoustic modes become confined. At $v = 1$ and 3.42 THz., the transverse optical modes of SnTe and PbTe are real respectively. Hence we observe a single line at those frequencies. We have thus understood the nature of the different physical solutions obtained in the different regions of the phonon and periodon dispersion in bulk and superlattice structure of IV–VI semiconductors. This type of study in superlattices will help in the understanding of various electronic and vibrational properties.

Acknowledgement

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