

Phonon States of Crystalline Film-Structures

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Abstract

The phonon spectra and states in three-dimensional bounded structures - ideal crystal films were studied as continuation of the investigation on analogous one- and two-dimensional systems. The comparison to corresponding unbounded crystal structures was done. Solving the Hamilton's equation of motion, the system of homogeneous difference equations for determination of the amplitude of small mechanical disturbances which extended through the observed system, was formulated. From the condition of nontriviality of the solutions of this system of equations, the phonon dispersion law with main characteristics - appearance of energy gap, was obtained. In the model structure, the acoustic phonon branches of optical type arise as the exclusive consequence of finite boundaries of system. The gap magnitude determining the different thermodynamical behaviour of film-structures, decreases with increase of film thickness and disappears with its increase.

1. Introduction

The scope of our study in this paper is limited to the analysis of the phonons, i.e. phonon behaviour in thin layered structures or crystalline films, which implies the existence of two boundary surfaces perpendicular to a preferred direction. Besides that, these film-structures could be doped by foreign atoms from one or both sides of the boundary surfaces [1,2] in which way the internal configuration of the atom distribution is disturbed [3,4].

Since elastic constants and atomic masses define phonon spectra and states, we conclude that they must be different in the film-structures with respect to the corresponding ones in the ideal unbounded and translationally invariant crystalline structures [5,6].

We shall study the thin film "cut-out" from the ideal cubic crystalline structure with lattice constants $a_x = a_y = a_z = a$. This structure has a finite width in the z -direction, while XY -planes are assumed to be infinite, meaning that the structure possesses two infinite boundary surfaces (parallel to the unbounded XY -planes) lying at $z = 0$ and $z = L$ (Fig.1.1). The number of the atoms located along z -direction is assumed to be N_z , and it is also assumed that torsion constants $C_{\alpha\beta}$ ($\alpha \neq \beta$) can be neglected with respect to the elongation constants $C_{\alpha\alpha}$ [7,8]. These structures will be entitled the ideal crystalline films.

The starting point of our study will be the standard Hamiltonian of the phonon system [6-9] in the nearest neighbour approximation:

$$H_{ID} = \frac{1}{2} \sum_{\vec{n}} \frac{p_{\vec{n}}^2}{M_{\vec{n}}} + \frac{1}{4} \sum_{\vec{n}, \vec{\lambda}} C_{\vec{n}, \vec{\lambda}} (\vec{u}_{\vec{n}} - \vec{u}_{\vec{n}+\vec{\lambda}})^2 \quad (1.1)$$

where: $\vec{p}_{\vec{n}}$ and $\vec{u}_{\vec{n}}$ - are the momentum and displacement of the atom of mass $M_{\vec{n}}$ at the crystal site $\vec{n} = a(n_x \vec{e}_x + n_y \vec{e}_y + n_z \vec{e}_z)$, while $C_{\vec{n}, \vec{\lambda}} \equiv C_{\vec{\lambda}, \vec{n}}$ - is Hooke's elastic constants between the atom at the site \vec{n} and its neighbouring atoms at the site $\vec{m} = \vec{n} + \vec{\lambda}$, $\vec{\lambda} = a(\vec{e}_x + \vec{e}_y + \vec{e}_z)$.

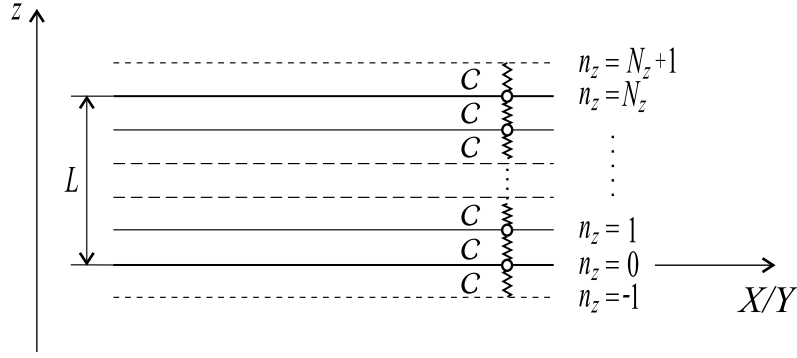


Figure 1.1: Schematic representation of the models of ideal crystalline film-structures

2. Normal frequencies and normal modes of phonons in ideal films

The Hamiltonian of the phonon subsystem of the model film-structure in the nearest neighbours approximation [7-10] is given in the form (1.1), where:

$$-\frac{N_\beta}{2} \leq n_\beta \leq \frac{N_\beta}{2}; \quad N_\beta \sim 10^8; \quad \beta \in (x, y); \quad 0 \leq n_z \leq N_z; \quad N_z = \frac{L}{a} \sim 20.$$

The concept of the ideal film means here the model of the crystal bounded by two parallel surfaces which can "breathe" (no rigid walls) along one crystallographic direction (which we choose for the positive direction of z -axis) perpendicular to the boundary surfaces and unbounded in the two other remaining directions. Furthermore, besides boundaries, there are no other defects in the ideal film, so inside the boundaries we encounter single atom tetragonal structure.

The Hamiltonian (2.1) adapted to the above mentioned model can be separated into two parts: the first one H_S , which includes "surface" terms and the second one H_B , which includes "bulk" terms subject to the conditions (see Fig.1):

$$M_{n_x, n_y, n_z} \equiv M; \quad C_{n_x n_y n_z; n_x \pm 1, n_y n_z}^\alpha = C_{n_x n_y n_z; n_x n_y \pm 1, n_z}^\alpha = C_{n_x n_y n_z; n_x n_y n_z \pm 1}^\alpha = C_\alpha.$$

Since there are no layers for $n_z \leq -1$ and for $n_z \geq N_z + 1$, we must include the following condition, too:

$$u_{n_x, n_y, l}^\alpha = 0; \quad l \leq -1 \wedge l \geq N_z + 1 \quad (\text{i.e. } l \notin [0, N_z]).$$

If we would assign $C_{-1}^\alpha = C_{N_z+1}^\alpha = 0$, then the boundary atoms (for $n_z = 0$ and $n_z = N_z$) would be "frozen", i.e. we would have the effects of rigid walls [11]. In this way, the expression for the total Hamiltonian of the ideal crystalline film obtains the following form:

$$H_{IF} = H_S + H_B, \quad (2.1)$$

where:

$$\begin{aligned}
H_S &= \frac{1}{2M} \sum_{\alpha} \sum_{n_x, n_y} \left[\left(p_{n_x, n_y, 0}^{\alpha} \right)^2 + \left(p_{n_x, n_y, N_z}^{\alpha} \right)^2 \right] + \\
&+ \frac{1}{4} \sum_{\alpha} \mathcal{C}_{\alpha} \sum_{n_x, n_y} \left[2 \left(u_{n_x, n_y, 0}^{\alpha} \right)^2 + 2 \left(u_{n_x, n_y, N_z}^{\alpha} \right)^2 + \right. \\
&+ \left(u_{n_x, n_y, N_z-1}^{\alpha} - u_{n_x, n_y, N_z}^{\alpha} \right)^2 + \left(u_{n_x, n_y, 1}^{\alpha} - u_{n_x, n_y, 0}^{\alpha} \right)^2 + \\
&+ \left(u_{n_x, n_y, 0}^{\alpha} - u_{n_x+1, n_y, 0}^{\alpha} \right)^2 + \left(u_{n_x, n_y, 0}^{\alpha} - u_{n_x-1, n_y, 0}^{\alpha} \right)^2 + \\
&+ \left(u_{n_x, n_y, 0}^{\alpha} - u_{n_x, n_y+1, 0}^{\alpha} \right)^2 + \left(u_{n_x, n_y, 0}^{\alpha} - u_{n_x, n_y-1, 0}^{\alpha} \right)^2 + \\
&+ \left(u_{n_x, n_y, N_z}^{\alpha} - u_{n_x+1, n_y, N_z}^{\alpha} \right)^2 + \left(u_{n_x, n_y, N_z}^{\alpha} - u_{n_x-1, n_y, N_z}^{\alpha} \right)^2 + \\
&+ \left. \left(u_{n_x, n_y, N_z}^{\alpha} - u_{n_x, n_y+1, N_z}^{\alpha} \right)^2 + \left(u_{n_x, n_y, N_z}^{\alpha} - u_{n_x, n_y-1, N_z}^{\alpha} \right)^2 \right] ;
\end{aligned} \tag{2.2}$$

$$\begin{aligned}
H_B &= \frac{1}{2M} \sum_{\alpha} \sum_{n_x, n_y} \left(p_{n_x, n_y, n_z}^{\alpha} \right)^2 + \frac{1}{4} \sum_{\alpha} \mathcal{C}_{\alpha} \times \\
&\times \sum_{n_x, n_y} \left\{ \sum_{n_z=1}^{N_z-1} \left[\left(u_{n_x+1, n_y, n_z}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 + \left(u_{n_x-1, n_y, n_z}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 + \right. \right. \\
&+ \left. \left(u_{n_x, n_y+1, n_z}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 + \left(u_{n_x, n_y-1, n_z}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 \right] + \\
&+ \left. \sum_{n_z=2}^{N_z-2} \left[\left(u_{n_x, n_y, n_z+1}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 + \left(u_{n_x, n_y, n_z-1}^{\alpha} - u_{n_x, n_y, n_z}^{\alpha} \right)^2 \right] + \right\} .
\end{aligned} \tag{2.3}$$

We have decided to use the approach of canonical equations of motion [6-9] for the determination of possible frequencies (energy spectrum) and the states of phonons. We start from the following system of the equations of motion for the phonon displacements:

- for $n_z = 0$:

$$\begin{aligned}
\ddot{u}_{n_x, n_y, 0}^{\alpha} &- \Omega_{\alpha}^2 \left(u_{n_x+1, n_y, 0}^{\alpha} + u_{n_x-1, n_y, 0}^{\alpha} + u_{n_x, n_y+1, 0}^{\alpha} + \right. \\
&+ \left. u_{n_x, n_y-1, 0}^{\alpha} + u_{n_x, n_y, 1}^{\alpha} - 6u_{n_x, n_y, 0}^{\alpha} \right) = 0 ,
\end{aligned} \tag{2.4}$$

- for $1 \leq n_z \leq N_z - 1$:

$$\begin{aligned}
\ddot{u}_{n_x, n_y, n_z}^{\alpha} &- \Omega_{\alpha}^2 \left(u_{n_x+1, n_y, n_z}^{\alpha} + u_{n_x-1, n_y, n_z}^{\alpha} + u_{n_x, n_y+1, n_z}^{\alpha} + \right. \\
&+ \left. u_{n_x, n_y-1, n_z}^{\alpha} + u_{n_x, n_y, n_z+1}^{\alpha} - u_{n_x, n_y, n_z-1}^{\alpha} - 6u_{n_x, n_y, n_z}^{\alpha} \right) = 0 ,
\end{aligned} \tag{2.5}$$

- for $n_z = N_z$:

$$\begin{aligned}
\ddot{u}_{n_x, n_y, N_z}^{\alpha} &- \Omega_{\alpha}^2 \left(u_{n_x+1, n_y, N_z}^{\alpha} + u_{n_x-1, n_y, N_z}^{\alpha} + u_{n_x, n_y+1, N_z}^{\alpha} + \right. \\
&+ \left. u_{n_x, n_y-1, N_z}^{\alpha} + u_{n_x, n_y, N_z-1}^{\alpha} - 6u_{n_x, n_y, N_z}^{\alpha} \right) = 0 ,
\end{aligned} \tag{2.6}$$

where $\Omega_{\alpha} = \sqrt{\mathcal{C}_{\alpha}/M}$. The solution of this system of N_z+1 homogeneous differential-difference equations for phonon displacements can be looked for in the form of the product of an unknown

function (along z -axis) and harmonic function of the position (within XY -plane) known from the bulk solutions , i.e.

$$u_{n_x, n_y, n_z}^\alpha(t) = \sum_{k_x, k_y} \sum_{k_z} \int_{-\infty}^{+\infty} d\omega e^{ia(k_x n_x + k_y n_y) - it \omega} \Phi_{n_z}^\alpha ; \quad \Phi_{n_z}^\alpha \equiv \Phi_{n_z}^\alpha(k_z, \omega) . \quad (2.7)$$

Substituting this expression into the equations (2.5-7) we obtain:

$$\begin{aligned} R \Phi_0^\alpha + \Phi_1^\alpha &= 0 \\ \Phi_0^\alpha + R \Phi_1^\alpha + \Phi_2^\alpha &= 0 \\ \cdot &\cdot \\ \Phi_{n_z-1}^\alpha + R \Phi_{n_z}^\alpha + \Phi_{n_z+1}^\alpha &= 0 \\ \cdot &\cdot \\ \Phi_{N_z-2}^\alpha + R \Phi_{N_z-1}^\alpha + \Phi_{N_z}^\alpha &= 0 \\ \Phi_{N_z-1}^\alpha + R \Phi_{N_z}^\alpha &= 0 , \end{aligned} \quad (2.8)$$

where:

$$R \equiv W_\alpha^2 - 4 \mathcal{F}_{k_x k_y} - 2 ; \quad W_\alpha \equiv \frac{\omega}{\Omega_\alpha} ; \quad \mathcal{F}_{k_x k_y} \equiv \sin^2 \frac{ak_x}{2} + \sin^2 \frac{ak_y}{2} . \quad (2.9)$$

In this way the system of $N_z + 1$ differential - difference equations (2.5 – 7) turns into a system of $N_z + 1$ homogeneous algebraic-difference equations (2.9). In order that this system possesses nontrivial solutions, its determinant:

$$\mathcal{D}_{N_z+1}(R) = \begin{vmatrix} R & 1 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 \\ 1 & R & 1 & 0 & \cdots & 0 & 0 & 0 & 0 \\ 0 & 1 & R & 1 & \cdots & 0 & 0 & 0 & 0 \\ \cdot & \cdot & \cdot & \cdot & \ddots & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 0 & 0 & 0 & 0 & \cdots & 1 & R & 1 & 0 \\ 0 & 0 & 0 & 0 & \cdots & 0 & 1 & R & 1 \\ 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 1 & R \end{vmatrix} \quad (2.10)$$

must vanish. The roots (poles) of this determinant represent one of the forms of Chebishev's polynomials of the second order [11-14] and can be written in the form:

$$\mathcal{D}_{N_z+1}(R) = \frac{\sin[(N_z + 2) \xi]}{\sin \xi} ; \quad \xi \neq 0 , \quad (2.11)$$

where: $R = 2 \cos \xi$. Above mentioned condition ($\mathcal{D}_{N_z+1}(R_\nu) = 0$) is satisfied for:

$$\xi_\nu = \frac{\pi \nu}{N_z + 2} ; \quad \nu = 1, 2, 3, \dots , N_z + 1 , \quad (2.12)$$

whose substitution into (2.10) leads to the expression for demanded (possible) unknown phonon frequencies:

$$\omega_{k_x k_y}^\alpha(\mu) = 2 \Omega_\alpha \sqrt{\mathcal{G}_\mu^I + \mathcal{F}_{k_x k_y}} , \quad (2.13)$$

where:

$$\mathcal{G}_\mu \equiv \sin^2 \frac{ak_z(\mu)}{2} ; \quad k_z(\mu) = \frac{\pi}{a} \frac{\mu}{N_z + 2} ; \quad \mu \equiv N_z + 2 - \nu = 1, 2, 3, \dots , N_z + 1 . \quad (2.14)$$

One must notice that contrary to k_x and k_y which range from 0 to π/a , one has:

$$k_z^{min} \equiv k_z(1) = \frac{\pi}{a} \frac{1}{N_z + 1} > 0; \quad k_z^{max} \equiv k_z(N_z + 1) = \frac{\pi}{a} \frac{N_z + 1}{N_z + 2} < \frac{\pi}{a}, \quad (2.15)$$

because $N_z \ll (N_x, N_y)$.

If one divides the system of equations (2.9) by $\Phi_0^\alpha \equiv \Phi_0^\alpha(k_z)$ and rejects the last equation, this system is obtained in the new form:

$$\begin{aligned} R_\nu + \varrho_1 &= 0, & \text{for: } n_z &= 0; \\ 1 + R_\nu \varrho_1 + \varrho_2 &= 0, & \text{for: } n_z &= 1; \\ \varrho_{n_z-1} + R_\nu \varrho_{n_z} + \varrho_{n_z+1} &= 0, & \text{for: } 2 \leq n_z &\leq N_z - 1, \end{aligned} \quad (2.16)$$

where:

$$\varrho_{n_z} \equiv \varrho_{n_z}^\alpha = (\Phi_0^\alpha)^{-1} \Phi_{n_z}^\alpha \implies \Phi_{n_z}^\alpha = \Phi_0^\alpha \varrho_{n_z}^\alpha. \quad (2.17)$$

The last of the equations (2.17) is satisfied for:

$$\varrho_{n_z} = (-1)^{n_z} \{P \sin(n_z \xi_\nu) + Q \sin[(n_z - 1) \xi_\nu]\}, \quad (2.18)$$

and using this and (2.12) it follows:

$$\varrho_1 = -P \sin(\xi_\nu); \quad \varrho_2 = P \sin(2 \xi_\nu) + Q \sin(\xi_\nu).$$

Substituting these expressions into the first and second equation in the system (2.17) we arrive to the unknown coefficients $P \equiv P_\nu = R_\nu \sin^{-1} \xi_\nu$ and $Q \equiv Q_\nu = -\sin^{-1} \xi_\nu$, while returning them into (2.19), and (2.18), it follows:

$$\Phi_{n_z}^\alpha(k_z) = (-1)^{n_z} \frac{\sin[(n_z + 1) \xi_\nu]}{\sin \xi_\nu} \Phi_0^\alpha. \quad (2.19)$$

According to above calculations (combining (2.8), (2.20) and standard normalization [12]), one can easily obtain the final expression for phonon displacements in the form:

$$u_{n_x, n_y, n_z}^\alpha(t) = \sum_{k_x k_y} \sum_{\mu=1}^{N_z+1} \mathcal{N}_{n_z}^\alpha(k_x k_y, \mu) e^{ia(k_x n_x + k_y n_y) - it \omega_{k_x k_y}^\alpha(\mu)} \sin[(n_z + 1) a k_z(\mu)]; \quad (2.20)$$

$$\mathcal{N}_{n_z}^\alpha(k_x k_y, \mu) = (-1)^{n_z} \sqrt{\frac{\hbar}{MN_x N_y (N_z + 2) \omega_{k_x k_y}^\alpha(\mu)}}.$$

Comparing the result obtained here with the corresponding one for ideal infinite structures, one can conclude that mechanical vibrations in the ideal unbounded structure are plane waves in all spatial directions, while in the thin film they represent the superposition of the standing waves in z -direction and plane waves in XY -planes. It is also evident that the displacement amplitude in the films is $\sim 10^4 \sqrt{2/N_z}$ times larger¹ than the amplitude in corresponding unbounded structures.

Using (2.14) one can determine the dispersion law for phonons in thin undeformed ideal film²:

$$E_{k_x k_y}^\alpha(\mu) \equiv \hbar \omega_{k_x k_y}^\alpha(\mu) = E_\alpha \sqrt{\mathcal{G}_\mu + \mathcal{F}_{k_x k_y}}, \quad (2.21)$$

¹For very thin films $N_z \sim 20$, so the factor of the amplitude increase can achieve even 10^3 .

²Most common treatment is that using classical procedure, for example, second quantization method [14], on the basis of (2.2-4), (2.15) and (2.21), the Hamiltonian H_{IF} is diagonalized, and then the energy spectrum in the form (2.22) is readily obtained.

where $E_\alpha = 2\hbar\Omega_\alpha$ and which is valid together with (2.10) and (2.15). We represent graphically this energy spectrum at the Figure 2.1a) in function of XY-plane vector $k^2 = k_x^2 + k_y^2$: $\mathcal{E}_\mu^z \equiv \left(E_{k_x k_y}^z(\mu)/E_z\right)^2 = \mathcal{E}_{k_z(\mu)}^z \left(\mathcal{F}_{k_x k_y}\right)$, and the Figure 2.1b) shows the same spectrum in terms of z -axis vector $k_z = k_z(\mu)$: $\mathcal{E}_k^z \equiv \left(E_{k_x k_y}^z(\mu)/E_z\right)^2 = \mathcal{E}_{k_x k_y}^z \left(\mathcal{G}_\mu\right)$.

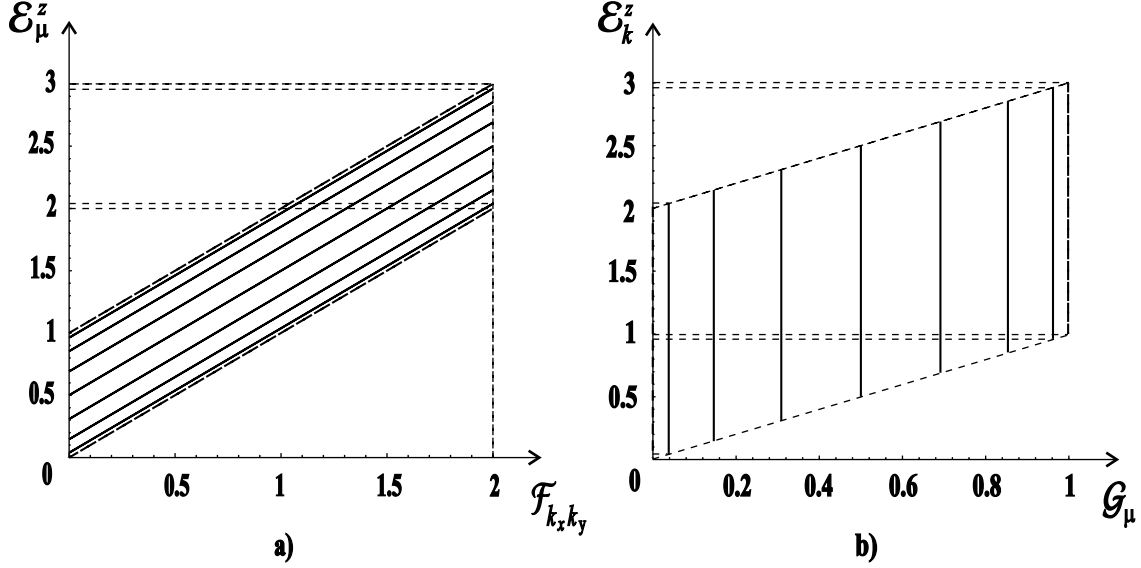


Figure 2.1: Energy spectrum of phonons in the ideal (ultrathin $N_z = 6$) crystalline films in function of two-dimensional (XY planar) wave vector and perpendicular wave vector. Within the band of bulk energies with continual spectrum (bulk limits are denoted by solid dashes lines) one can notice (7) allowed distance photon energies in the film studied (thin solid lines). One can notice the narrowing of the energy band and the existence of the energy gap.

One can clearly see from the plot explicit discreteness of the allowed energy levels of phonons in the ideal film with respect to the continuum of these values for the corresponding bulk-structures. All three acoustic frequencies in bulk-structures vanish when three-dimensional (spatial) vector $k = |\vec{k}|$ vanishes, while the minimal frequencies of phonons in the thin ideal film-structure are:

$$\omega_\alpha^{min} \equiv \omega_\alpha(k_x = k_y = 0, k_z = k_z^{min}) \approx \Omega_\alpha \frac{\pi}{N_z + 2} > 0. \quad (2.22)$$

On the other hand, maximal values of the frequencies of acoustic branches in the ideal infinite crystal tend to the value $(\omega_\alpha^B)_{max} = 2 \Omega_\alpha \sqrt{3}$ when $k_\alpha \rightarrow \pi/a$ ($\alpha = x, y, z$), while in the studied ideal film they are:

$$\omega_\alpha^{max} \equiv \omega_\alpha(k_x = k_y = \pi/a, k_z = k_z^{max}) \approx 2 \Omega_\alpha \sqrt{3} \left[1 - \frac{\pi^2/12}{(N_z + 2)^2} \right] < (\omega_\alpha^B)_{max}. \quad (2.23)$$

It can be also seen from the same figure that the width of the energy band in the film is narrower. From (2.23) and (2.24) we can determine the total narrowing of the band of allowed energies of the phonons in the film-structures with respect to the bulk band:

$$\mathcal{W}_\alpha \equiv \hbar \left[(\omega_\alpha^B)_{max} - \left(\omega_\alpha^{max} - \omega_\alpha^{min} \right) \right] \approx \hbar \Omega_\alpha \frac{\pi (N_z + 3)}{(N_z + 2)^2} > 0. \quad (2.24)$$

3. Conclusion

Studying and comparing the phonon spectra and states in the ideal unbounded and nondeformed (bulk) structures and the structures with broken translational symmetry (films) we have reached the following conclusions.

1. Mechanical vibrations in bulk structures are plane waves in all directions, while in the films they represent the superposition of the standing waves in z -directions (perpendicular to the boundary surfaces) and plane waves in XY -planes (parallel to boundary surfaces).
2. The amplitude of phonon displacements in the films depends on the film width and it is $\sim 10^4 \sqrt{2/N_z}$ times higher than in the ideal structures. This indicates their larger elastic "maneuvering space" without any negative effect to the mechanical properties of the given material (for example no breaking of interatomic bonds) which leads to higher resistance and higher melting point of the films with respect to bulk samples.
3. All three acoustic frequencies in bulk structures vanish for $\vec{k} \rightarrow 0$, while in the films they tend toward some minimal value depending on the film width. This means that phonons in the films possess the energy gap, that for their excitation (creation) one should spend certain energy, i.e. heat them up to certain - activation temperature, meaning that the system up to that temperature behaves as the "frozen" one, as if the phonons were not present.
4. Phonon gap, besides depending on the film width, depends also on the type of the atoms and their distribution along z -direction and also on the stoichiometric relation of the atoms injected in the films.

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