

**QUANTIZATION OF ENERGY IN 1D MODEL OF CRYSTAL
LATTICE WITH LOCAL PERTURBATIONS INDUCED
BY ION-BEAM IMPACT**

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Abstract

In this paper, we propose theoretical basis for investigation of dynamics of acoustic phonons in a thin layers containing nano-scale structural inhomogeneities. One-dimensional (1D) model of a crystal lattice was considered to reveal specific features of the processes arising in such system of phonons in equilibrium state. Standard quantization of energy of 1D ionic chain vibrating by acoustic frequencies was carried out while the presence of foreign ions in this chain was taken into account. Since only two dimensions are dominant in thin layers, only longitudinal vibrations of the chain in the plane of the layer were considered. Results showed that foreign ions affect the energy quantization. Phonon-phonon interaction between two phonon`s modes can be expected if the mass of foreign ions implanted by ion-beam differs from the mass of ions in the initial layer.

We believe that the obtained results will help to understand the character of phonon systems in nanostructured thin layers prepared by ion-bem technology, and will allow better explain some thermal and electrical phenomena associated with lattice dynamics in such layers.

Key words

acoustic phonon, quantization, thin layer, 1D ionic chain, phonon modes, structural inhomogeneities

1. INTRODUCTION

The study of the dynamics of phonon systems is one of the central tasks in the solid state physics. Quantization of energy of vibrating lattice described by phonons concept, much like quantization of electrons or holes, affects the electrical, optical and thermal properties of ultra-thin films and nanostructures. The consequences of phonon quantization can be found in the non-radiational and radiational relaxation of electrons (1–3), phonon bottleneck effect (4, 5) and stimulated far-infrared emission from quantum dots (6). Acoustic phonons play an

important role in the electrical and thermal properties of semiconductor materials. They are the dominant heat carriers in moderately doped semiconductors in a wide range of temperatures from cryogenic to almost the melting point (7). More recently it has been shown theoretically that confinement-induced modification of the acoustic phonon spectrum in free-surface ultrathin films (8) and nanowires (9–11) leads to a significant decrease of the in-plane lattice thermal conductivity even at room temperature. The decrease of thermal conductivity owing to phonon confinement may bear important consequences for the electronic industry in a view of continuous miniaturization. Folded acoustic phonons in layered medium have been studied theoretically by Rytov (12). Later, the folded phonons have been observed experimentally in quantum well superlattices (13). Many theoretical results for quantized phonons in free standing films, nanowires and spherical quantum dots have been obtained using the analogy with acoustics and classical mechanics (14–16).

The phonon concept valids when the anharmonic contribution in crystal vibration is small compared with the harmonic. In such a case, the different phonon modes are independent and do not interact with each other. However, this simple picture is only an approximation. In the case of large atom displacements, the higher-order terms obtained by expanding of the potential energy should be taken into account. Keeping the cubic term, the interaction between phonon modes can be described and thus the phonon-phonon interaction between two phonon`s modes can be explicitly considered (17).

In this contribution, we discuss the quantization of energy in 1D crystal lattice with defects in the long-wave approximation. The following two cases are considered:

1. The simplest example of a local perturbation of ion`s mass when the foreign ions are included to 1D ionic chain. In such a case, a discontinuity of stresses in the chain can be generated and vibration of the chain can be influenced by these local mass defects.
2. Perturbation of elastic moduli in the 1D crystal lattice. In such a case, we have a discontinuity of inter-atomic bonds. The interaction between two ions in the chain is characterized by the elastic force constant β which varies along the chain.

When talking about a local perturbation in a linear elastic medium or 1D crystal lattice, we mean the presence of foreign ions in ionic chain in the area of a few atomic distances, the elastic properties of which distinguish from those in the rest of the chain. There is a number of types of local and planar defects in real crystals. In the continuum theory, the presence of such defects in an elastic medium is associated with special boundary conditions at some interfaces for the equation of motion of the non-perturbated medium. Changes of mass and elastic constant of medium play a crucial role. For example in ref. (18), the boundary conditions consist of a local perturbation of the elastic moduli at the interface.

In this paper, we present a simple model for investiagtion of influencing local perturbations in crystal on the crystal lattice dynamics. The aim of the paper is to lay the foundation for a detailed qualitative analysis of the phonon system in locally perturbed media. We aim to identify respective contributions of local defects induced by ion-beam impact to the acoustic phonons energy distribution. Our attention is concentrated on the dynamics of equilibrium phonons, particularly on the spectral evolution of their distribution and its dependence on local changes of mass and elastic force constant.

2. VIBRATION OF NON-LINEAR ELASTIC CONTINUUM

Before analysis of behaviour of non-linear vibrating system, it is first necessary to analyze the linear system. As a classical example may be mentioned Hamiltonian of finite-dimensional system with coupled degrees of freedom. The behaviour of such system for small excitations is

determined by the form of the quadratic part of its Hamiltonian. This method corresponds to the linear dynamics.

Example of nonlinear systems, the behaviour of which depends on the linear part, is weakly interacting with dispersive waves in continuous media. These systems studied in wave turbulence often have analogs among discrete systems, e.g., a chain of coupled oscillators. Correspondingly, their quadratic Hamiltonians have analogs among the Galin–Arnold canonical forms (19). Theory of wave turbulence is very often applied to homogeneous systems, and the quadratic Hamiltonian describing linear interactions in such homogeneous systems is given by (20):

$$H = \int \left(A_{\vec{k}} |a_{\vec{k}}|^2 + \frac{1}{2} (B_{\vec{k}} a_{\vec{k}} a_{-\vec{k}} + B_{\vec{k}}^* a_{\vec{k}}^* a_{-\vec{k}}^*) \right) d\vec{k} , \quad [1]$$

where $a_{\vec{k}}$ is a complex-valued field.

By means of linear canonical transformation to the new variables $b_{\vec{k}}$, the mentioned Hamiltonian can be rewritten to the next normal form :

$$H = \int \omega_{\vec{k}} |b_{\vec{k}}|^2 d\vec{k} , \quad [2]$$

where $\omega_{\vec{k}}$ obeys a linear dispersive relationship.

The problem of searching the canonical form for the quadratic Hamiltonian for the inhomogeneous systems can be solved by the extension of certain oscillatory members onto the continuous space. Such a formulation lays down a necessary framework for generalization of the Hamiltonian description onto the inhomogeneous media. General Hamiltonian for the system of linear waves propagating in the inhomogeneous media for the variable $a_{\vec{q}}$ is given by the following quadratic form (21):

$$H = \int \left(A(\vec{q}, \vec{q}') a_{\vec{q}} a_{\vec{q}'}^* + \frac{1}{2} (B(\vec{q}, \vec{q}') a_{\vec{q}} a_{-\vec{q}'} + B^*(\vec{q}, \vec{q}') a_{\vec{q}}^* a_{-\vec{q}'}^*) \right) d\vec{q} d\vec{q}' . \quad [3]$$

In this paper, we try to point out analogies of our results obtained using discrete model with the above mentioned results [1-3]. It can be assumed that some nonlinearities in lattice vibration can be generated by the local structure perturbations.

2. 1D MODEL OF CRYSTAL LATTICE WITH LOCAL PERTURBATION

The 1D ion chain model of crystal lattice with local perturbation is studied (Fig.1). A 1D lattice seems to be an appropriate model that could, in addition, it allows for some mathematical treatment and thus a better theoretical understanding of the phenomena and mechanisms at play. Indeed, many mathematical results are known about the behaviour of waves in 1D lattices, concerning the existence of localized waves (22, 23), the form of those waves in the high-energy limit (24) or in the low-energy limit (25), or the behaviour under shock (26, 27).

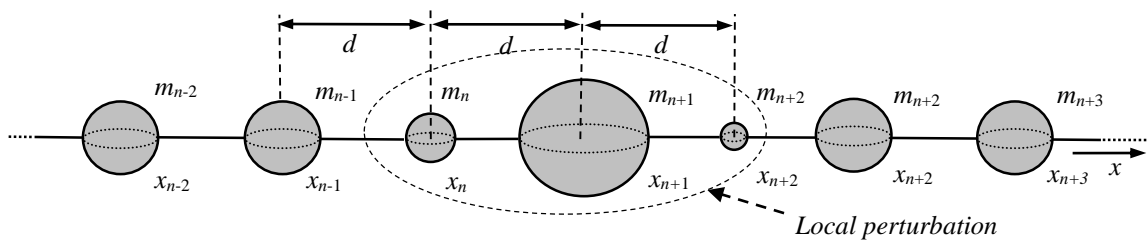


Fig.1 1D model of the crystal lattice. All ions in the chain are identical except ions in positions $n, n+1$ and $n+2$

Consider N ions of unequal masses $\{m_n\}$ and charges $\{Q_n\}$ with nonlinear nearest-neighbour interactions, described by a potential energy U . The position of the n -th ion will be denoted by x_n , where the ions are numbered in order of increasing value of their axial positions, so that $n > m$ implies that $x_n > x_m$ (see Fig.1). Initially, the particles are at rest at positions $\{x_{n0}\}$, which is an equilibrium state for the system.

Ions are bound relatively strongly in their equilibrium positions in the crystal lattice. The following analysis is based on the classical description of the dynamics of ions in the 1D crystal lattice in harmonic approximation.

4. EQUATIONS OF MOTION

We consider one-dimensional crystal represented by a linear chain of N ions (Fig.1), which is not inserted into the external field. It turns out that the results provided by the solution of such a simplified one-dimensional model can be applied in the real three-dimensional case. The total potential energy U of the chain depends on the positions of all ions $\{x_n\}$ in the chain, and in case of Coulomb interaction, it can be expressed by:

$$U = U(\{x_i\}) = \frac{1}{2} \sum_j \sum_k \frac{Q_j Q_k}{4\pi\epsilon_0 |x_j - x_k|}, \quad [4]$$

where Q_j and Q_k are the electric charges of the j -th and k -th ion respectively. Let $\{z_i\}$ are displacements of ions from equilibrium positions, displacement of n -th ion in the chain is defined as follows:

$$\tilde{z}_n = x_n - x_{n0}. \quad [5]$$

If displacements of ions from the equilibrium are small $\tilde{z}_n \ll d$, the potential energy U can be expanded to the Taylor series:

$$U = U(\{x_{i0}\}) + \sum_n \left(\frac{\partial U}{\partial x_n} \right)_{x_n=x_{n0}} \tilde{z}_n + \frac{1}{2!} \sum_n \sum_k \left(\frac{\partial^2 U}{\partial x_n \partial x_k} \right)_{\substack{x_n=x_{n0} \\ x_k=x_{k0}}} \tilde{z}_n \tilde{z}_k + \dots \quad [6]$$

and harmonic approximation can be applied, i.e. only the second degree polynomial function $U(\{\tilde{z}_i\})$ can be considered. Moreover, given that:

$$\left(\frac{\partial U}{\partial x_n} \right)_{x_n=x_{n0}} = 0, \quad [7]$$

due to the fact that potential energy U has a minimum for each equilibrium positions $\{x_{i0}\}$ (see Fig. 2), the potential energy U in the harmonic approximation is given by:

$$U = U_0 + \frac{1}{2} \sum_n \sum_s A_{n,s} \tilde{z}_n \tilde{z}_s, \quad \text{where } U_0 = U(\{x_{i0}\}) \text{ and } A_{n,s} = \left(\frac{\partial^2 U}{\partial x_n \partial x_s} \right)_{\substack{x_n=x_{n0} \\ x_s=x_{s0}}}. \quad [8]$$

Then the force exerted on the n -th ion in chain can be written as:

$$f_n = -\frac{\partial U}{\partial z_n} = -A_{n,n} \tilde{z}_n - \frac{1}{2} \sum_{i \neq n} A_{i,n} \tilde{z}_i - \frac{1}{2} \sum_{j \neq n} A_{n,j} \tilde{z}_j = -\sum_k A_{n,k} \tilde{z}_k. \quad [9]$$

If the displacement of all ions from equilibrium is the same ($\{\tilde{z}_i\} = \tilde{z}$), the whole crystal is shifted and force [9] acting on each ion in chain is zero in this case:

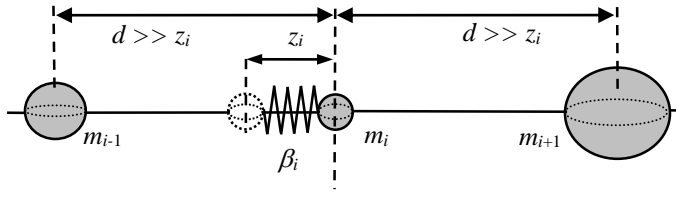


Fig. 2 Oscillation of ions in linear chain

$$-\sum_k A_{n,k} \tilde{z} = 0. \quad [10]$$

It results into the next condition for coefficients $A_{n,k}$ from the relation [10], subsequently:

$$\sum_k A_{n,k} = 0. \quad [11]$$

Assuming inhomogeneous 1D chain, there are different masses of

individual ions (see Fig.1) and also a variety of binding forces between them can be considered. Using the following notations:

$$A_{n,n-1} = -k_{n,n-1}, \quad A_{n,n+1} = -k_{n,n+1}, \quad A_{n,n} = k_{n,n} \quad [12]$$

and applying the tight binding approximation, the equation of motion for the n -th ion in the chain is given by:

$$f_n = m_n \frac{\partial^2 \tilde{z}_n}{\partial t^2} = k_{n,n-1} \tilde{z}_{n-1} - k_{n,n} \tilde{z}_n + k_{n,n+1} \tilde{z}_{n+1}. \quad [13]$$

From relation (11) it results that:

$$k_{n,n} = k_{n+1} + k_{n-1}, \quad [14]$$

and equation (13) can be rewritten to the next form:

$$m_n \frac{\partial^2 \tilde{z}_n}{\partial t^2} = k_{n,n-1} (\tilde{z}_{n-1} - \tilde{z}_n) + k_{n,n+1} (\tilde{z}_{n+1} - \tilde{z}_n). \quad [15]$$

System of equations of motion (15) for $n = 1 \dots N$ allows to describe the dynamics of ions in the frame of the established 1D chain model. Solution of this system is the set of functions $\{z_n(t)\}$, which describes time dependence of the displacements of ions from equilibrium in the chain.

5. ENERGY OF LINEAR CHAIN OF IONS

The total mechanical energy of linear chain of ions is determined by the sum of its kinetic energy W_k and potential energy U :

$$E = W_k + U. \quad [16]$$

Kinetic energy W_k is determined by:

$$W_k = \frac{1}{2} \sum_{n=1}^N m_n \dot{\tilde{z}}_n^2. \quad [17]$$

Potential energy U must be calculated by means of relation (8) in harmonic case and applying the tight binding approximation, only the impact of nearest neighbours is taken into account (such as in the case [13]).

In this case, we consider only interaction between the nearest neighbours of each ion, that means that only those members in the sum (8) are nonzero indexes of which are satisfying the conditions $s = n-1$, $s = n$ and $s = n+1$. Owing to (12) and (14), the potential energy of linear chain in the tight binding approximation can be written as:

$$U = U_0 + \left\{ -\frac{1}{2} \sum_n k_{n,n-1} \tilde{z}_n \tilde{z}_{n-1} + \frac{1}{2} \sum_n k_{n,n-1} \tilde{z}_n^2 + \frac{1}{2} \sum_n k_{n,n+1} \tilde{z}_n^2 - \frac{1}{2} \sum_n k_{n,n+1} \tilde{z}_n \tilde{z}_{n+1} \right\}. \quad [18]$$

Given the large number of ions in the chain, the solution of the problem changes only insignificantly if the index of each member in the last two sums in the right side of relation [18] will be reduced by one (i.e. $n \rightarrow n-1$). If we denote $k_{n,n-1} = \beta_n$, then the potential energy of the chain [18] is:

$$U = U_0 + \sum_n \frac{1}{2} \beta_n (\tilde{z}_n - \tilde{z}_{n-1})^2 \quad [19]$$

and the total energy can be determined by the formula:

$$E = \frac{1}{2} \sum_{n=1}^N m_n \dot{\tilde{z}}_n^2 + \sum_n \frac{1}{2} \beta_n (\tilde{z}_n - \tilde{z}_{n-1})^2. \quad [20]$$

Discrete Fourier transform (DFT) of the set of functions $\{z_n(t)\}$, ion masses $\{m_n\}$ and spring constants $\{\beta_n\}$ can be found in the next form:

$$\tilde{z}_n = \frac{1}{\sqrt{N}} \sum_j \left\{ a_j e^{iq_j n a} + a_j^* e^{-iq_j n a} \right\}, \text{ where: } a_j = \sqrt{N} A_j e^{i\omega_j t}, \quad a_j^* = \sqrt{N} A_j^* e^{-i\omega_j t}, \quad [21]$$

$$m_n = \sum_k c_k e^{-i\frac{2\pi}{N}nk}, \quad \beta_n = \sum_m d_m e^{-i\frac{2\pi}{N}nm} \quad [22]$$

for the reason of transformation of formula (20) to different representation. We expect, that from the Born-Karman boundary condition, it results:

$$q_j = \frac{2\pi}{Na} j, \quad j = 0, 1, 2, \dots \quad [23]$$

Since it is clear that:

$$\frac{da_j}{dt} = \dot{a}_j = i\omega_j a_j, \quad \frac{da_j^*}{dt} = \dot{a}_j^* = -i\omega_j a_j^* \quad [24]$$

after substituting the right side of relations [21] and [22] to the formula [17], the kinetic energy of the inhomogeneous chain can be determined as:

$$W_k = \frac{1}{2} \sum_j \sum_k c_k \left(-\omega_j \omega_{-(j-k)} a_j a_{-(j-k)} + \omega_j \omega_{j-k} a_j a_{j-k}^* + \omega_j \omega_{j+k} a_j^* a_{(j+k)} - \omega_j \omega_{-(j+k)} a_j^* a_{-(j+k)}^* \right). \quad [25]$$

Similarly, if we substitute the right side of relations (21) and (22) to the formula (19), the potential energy of the chain is:

$$U = \frac{1}{2} \sum_j \sum_m d_m \left\{ \left(a_j a_{-(j-m)} + a_j^* a_{(j-m)}^* \right) \left(1 - e^{-i\frac{2\pi}{N}j} \right) \left(1 - e^{i\frac{2\pi}{N}(j-m)} \right) + \left(a_j^* a_{j+m} + a_j a_{-(j+m)}^* \right) \left(1 - e^{i\frac{2\pi}{N}j} \right) \left(1 - e^{-i\frac{2\pi}{N}(j+m)} \right) \right\} \quad [26]$$

The total energy of vibrating ions in the chain can be determined as the sum of kinetic energy [25] and potential energy [26]:

$$E = \frac{1}{2} \sum_j \sum_k c_k \left(-\omega_j \omega_{-(j-k)} a_j a_{-(j-k)} + \omega_j \omega_{j-k} a_j a_{j-k}^* + \omega_j \omega_{j+k} a_j^* a_{(j+k)} - \omega_j \omega_{-(j+k)} a_j^* a_{-(j+k)}^* \right) + \frac{1}{2} \sum_j \sum_m d_m \left\{ \left(a_j a_{-(j-m)} + a_j^* a_{(j-m)}^* \right) \left(1 - e^{-i\frac{2\pi}{N}j} \right) \left(1 - e^{i\frac{2\pi}{N}(j-m)} \right) + \left(a_j^* a_{j+m} + a_j a_{-(j+m)}^* \right) \left(1 - e^{i\frac{2\pi}{N}j} \right) \left(1 - e^{-i\frac{2\pi}{N}(j+m)} \right) \right\}. \quad [27]$$

Taking into account:

$$c_k = \frac{1}{N} \sum_n m_n e^{i\frac{2\pi}{N}nk}, \quad \frac{1}{N} \sum_s e^{i\frac{2\pi}{N}sk} = \delta_{k,0} \quad \text{and} \quad \omega_{-(j-k)} = \omega_{(j-k)}, \quad \omega_{-(j+k)} = \omega_{(j+k)}$$

and after re-index sums, the above formula (27) can be formally rewritten as:

$$E = 2m \sum_j \omega_j^2 a_j a_j^* + 2m \sum_j \sum_k A_{jk} a_j a_j^* \omega_j \omega_{(j+k)} + \Omega, \quad [28]$$

$$\text{where: } \Omega = \frac{1}{2} \sum_j \sum_k D_k \left[2a_j^* a_{(j+k)} + a_{j+k} a_{-j} + a_j^* a_{-(j+k)}^* \right] \left(1 - e^{i\frac{2\pi}{N}j} \right) \left(1 - e^{-i\frac{2\pi}{N}(j+k)} \right) \quad [29]$$

$$\text{and: } A_{jk} = \frac{C_k}{4ma_j a_j^*} \left(a_{(j+k)} - a_{-(j+k)}^* \right) \left(a_j^* - a_{-j} \right).$$

[30]

Constants C_k and D_k are determined by means of the inverse discrete Fourier transform of ion masses $\{m_n\}$ and spring constants $\{\beta_n\}$:

$$C_k = \frac{1}{N} \sum_n m_n e^{i\frac{2\pi}{N}nk} - m\delta_{k,0}, \quad D_k = \frac{1}{N} \sum_n \beta_n e^{i\frac{2\pi}{N}nk} - \beta\delta_{k,0}, \quad \text{with: } m = \frac{1}{N} \sum_{n=1}^N m_n, \quad \beta = \frac{1}{N} \sum_{n=1}^N \beta_n. \quad [31]$$

We used here a simplified discrete 1D model of crystal for description of lattice vibrational energy. Some important features of acoustic waves can be analyzed on the basis of the result observed in case of such 1D polyatomic chain. The behaviour of energy of vibrating lattice can be studied. There is an apparent analogy of our results [28] with the formula [3]. Next, we can study the effects of structural inhomogeneities produced by the quantization of vibrational degrees of freedom in the crystal lattice. A straightforward extension of this methodology can be the calculation of vibrational excitations.

6. CONCLUSION

In order to study the effect of foreign ions on the acoustic phonon dynamics in linear chain of ions, we implemented quantization to the calculation of energy of the chain. The result [28] shows that the total energy of the vibrating lattice can be written as superposition of three contributions which correspond to the concept of phonons. The first of them is characteristic for unperturbed medium. The second and third contributions are generated by the presence of foreign ions in the chain. Matrix elements A_{jk} reflect the mass of foreign ions, and parameter Ω is related to the interatomic forces acting in the structure.

Next, we intend to investigate the mechanism of acoustical excitation in nanostructures and discuss the possible influence of local lattice defects on the phonon assisted phenomena. For calculation of different macroscopic characteristics of nanostructures, such as thermal and electrical conductivity, heat capacity, etc., it is required to know the spectral density of the phonon mode distribution, e.g. phonon density of states. It can be expected on the basis of our result that local inhomogeneities change the shape of the dispersion curves of phonons both quantitatively and qualitatively.

For example, the lattice thermal conductivity σ can be estimated using the kinetic theory of gases [28, 29]:

$$\sigma = \frac{1}{3} C v_{ph} L = \frac{1}{3} C v_{ph}^2 \tau,$$

where C is the heat capacity per unit volume, v_{ph} the mean sound velocity of the phonons, L the mean free path of the phonons, τ the phonon relaxation time. Just the thermal capacity C depends on the acoustic phonon modes as can be seen from Deby's theory. Therefore, C could be influenced by local perturbation of structure. Our results can be useful for understanding the contribution of the phonon dispersion in the thermal conductivity of ultra-thin layers, as well as in the design of efficient thermal management and thermoelectric devices.

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