A Theoretical Model of Thermal Conductivity for Multilayer Nitride-Based Nanosystems

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***Abstract*—A new generalized analytical theoretical model of thermal conductivity for multilayer nanostructures based on nitride semiconductors is proposed. The developed theory qualitatively complements and generalizes the investigations, the results of which were performed for one- or two-layer nanostructures. The theory has its mathematical basis for the application of another type of boundary conditions for the components of the phonon field and components of the stress tensor. The obtained results can be performed to describe the processes associated with the theoretical conductivity of nanostructures with an arbitrary number of layers in them. The proposed theoretical investigations are applied to the calculation of the thermal conductivity of experimentally created nanostructures.**

Keywords— thermal conductivity, acoustic phonons, nitride-based semiconductors, nanostructure, quantum cascade detector

I. Introduction

Nowadays, it is almost impossible to imagine semiconductor electronics without the application of nanomaterials and nanostructures of various geometric symmetry [1-4]. The complexity of describing physical phenomena in nanostructures with a large number of layers is very often inextricably associated with the use of completely new mathematical models and the need to establish the features of the components of the studied quantities (field components, displacement of the medium, etc.) on the heteroboundaries between the different media of these nanosystems.

For quantum cascade detectors based on nitride semiconductors GaN, AlN, GaAlN [2-4], in connection with the functional capabilities of the effective operation of these nanodevices in a wide temperature range, it is very important to clarify the aspects of the interaction of electrons with optical and acoustic phonons. piezoelectric effect, thermal conductivity [5-9].

The study of the thermal conductivity of nitride nanostructures is directly related to the calculation of the spectral characteristics of various types of acoustic phonons Ω_n and the corresponding group velocities υ_g. The direct mathematical model of acoustic phonons for a nanostructure with a crystal lattice of wurtzite or zinc blende is rather complicated. Therefore, in most of the existing papers, significant simplifications of theoretical models were carried out, which, as a result, can only qualitatively describe the real essence of physical processes. These simplifications include the following. First, in order to avoid problems with the use of a large number of boundary conditions, only idealized mono- and two-layer nanostructures with absolutely elastic walls were considered [7, 8]. Secondly, it was groundlessly postulated that the systems of differential equations describing phonon modes have only numerical solutions, which thus made it impossible to develop the analytical theory.

The analytical theory of acoustic phonons and electronic processes caused by them in plane resonant tunneling nanosystems has been developed quite recently [9]. With its use in the proposed paper, a theoretical model of the thermal conductivity of a cascade of a quantum cascade detector based on AlN/GaN is developed. The effect of partial phonon transmission at the boundaries of the nanostructure layers is taken into account. The results of the proposed theory are taken to perform calculations for the thermal conductivity of the experimentally investigated nanostructure.

II. A Mathematical Model of Thermal Conductivity for Multilayer Nanosystems

We will investigate the propagation of acoustic phonon waves and thermal conductivity of a multilayer nanostructure, the geometric scheme of which is schematically shown in Fig. 1.

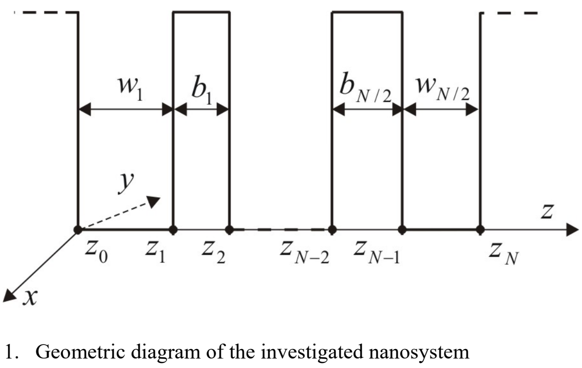

Fig. 1. Geometric diagram of the investigated nanosystem

First of all, given the fact that we investigate a multilayer nanosystem, we represent its density and elastic moduli in the following form:

$$\begin{bmatrix} \rho(z) \\ C_{\alpha\beta}(z) \end{bmatrix} = \sum_{p=1}^{N} \begin{bmatrix} \rho^{(p)} \\ C_{\alpha\beta}^{(p)} \end{bmatrix} \left[\theta(z_p - z_{p-1}) - \theta(z_p - z_{p+1})\right] \quad (1)$$

where $\rho^{(p)}$ - density of a nanostructure layer related to a GaN or AlN semiconductors, $C_{\alpha\beta}^{(p)}$ - similarly specified elastic constants of these materials (are represented in the two-index Voigt notation), $\theta(z)$ – Heaviside unit function.

To establish the spectral characteristics of phonons related to the acoustic energy range, we use the model of an elastic semiconductor medium. This leads to the need of finding solutions to the following equation:

$$\rho(z)\frac{\partial^2 u_l(r,t)}{\partial t^2} = \frac{\partial \sigma_{lk}(r)}{\partial x_k};\ l,\ k = (1;\ 2;\ 3) \quad (2)$$

where

$$\sigma_{ik}(r) = \frac{1}{2} C_{iklm}(z)\left(\frac{\partial u_l(r)}{\partial x_m} + \frac{\partial u_m(r)}{\partial x_l}\right),\ l,\ m = (1;\ 2;\ 3) \quad (3)$$

- is the stress tensor. Next, using relation (3) in equation (2), performing the transition $C_{iklm}(z) \to C_{\alpha\beta}(z)$ and taking into account the explicit form of the elastic constant tensor [5, 6, 9], we will look for solutions to this equation in the following form:

$$u_l(r,t) = \sum_{p=1}^{N} \left(u_1^{(p)}(z)\ u_2^{(p)}(z)\ u_3^{(p)}(z)\right)^T \times \\ \times \left[\theta(z_p - z_{p-1}) - \theta(z_p - z_{p+1})\right] e^{i(\omega t - qx)} \quad (4)$$

where the symbol "T" denotes a transposition operation. Now equation (2) takes the following matrix form:

$$M_i^{(p}(z)\ddot{u}_i^{(p)}(z) + \rho^{(p)}\omega^2 u_i^{(p)}(z) = 0;\ i = (1;\ 2;\ 3) \quad (5)$$

In equation (5), the matrix $M_i^{(p}(z)$ looks like [11, 13, 14]:

$$M_i^{(p}(z) = \begin{bmatrix} -C_{44}^{(p)}\frac{d^2}{dz^2} + C_{11}^{(p)} q^2 & 0 & iq\left(C_{31}^{(p)} + C_{44}^{(p)}\frac{d}{dz}\right) \\ 0 & -C_{44}^{(p)}\frac{d^2}{dz^2} + C_{66}^{(p)} q^2 & 0 \\ iq\left(C_{31}^{(p)} + C_{44}^{(p)}\frac{d}{dz}\right) & 0 & -C_{33}^{(p)}\frac{d^2}{dz^2} + C_{44}^{(p)} q^2 \end{bmatrix}. \quad (6)$$

In what follows, we will not consider transverse phonons with the component $u_2^{(p)}(z)$, which, as is known [5], do not contribute to the electron–phonon interaction and, accordingly, to the conductivity of the nanosystem. Then for the functions describing the displacements $u_1^{(p)}(z)$ and $u_3^{(p)}(z)$, the following self-consistent system of equations is obtained:

$$\begin{cases} -\dfrac{d^2 u_1^{(p)}(z)}{dz^2} + iqc_1\dfrac{du_3^{(p)}(z)}{dz} - k_1^2(q,\omega)u_1^{(p)}(z) = 0; \\ -\dfrac{d^2 u_3^{(p)}(z)}{dz^2} + iqc_3\dfrac{du_1^{(p)}(z)}{dz} - k_3^2(q,\omega)u_3^{(p)}(z) = 0, \end{cases} \quad (7)$$

where such notation was made to simplify the form of the equations:

$$c_1 = \frac{C_{31}^{(p)} + C_{44}^{(p)}}{C_{44}^{(p)}};\ k_1^2(q,\omega) = \frac{\rho^{(p)}\omega^2 - q^2 C_{11}^{(p)}}{C_{44}^{(p)}}; \\ c_3 = \frac{C_{31}^{(p)} + C_{44}^{(p)}}{C_{33}^{(p)}};\ k_3^2(q,\omega) = \frac{\rho^{(p)}\omega^2 - q^2 C_{44}^{(p)}}{C_{33}^{(p)}}. \quad (8)$$

Desired exact solutions analytical solutions ($u_1(z)$, $u_3(z)$) of the system of equations (7) are obtained in analytical form by using the method developed in [9]. For a complex nanosystem with a large number of layers, they can be presented as follows:

$$u_{1(3)}(q,\omega,z) = u_{1(3)}^{(0)}(q,\omega,z)\theta(-z) + u_{1(3)}^{(N+1)}(q,\omega,z)\theta(z - z_N) + \\ + \sum_{p=1}^{N} u_{1(3)}^{(p)}(q,\omega,z)\left[\theta(z - z_{p-1}) - \theta(z - z_p)\right], \quad (9)$$

where respectively:

$$u_1^{(p)}(q,\omega,z) = -iqc_1\left(\frac{\lambda_1}{\left\|\alpha_1^{(p)}\right\|}\left(A_1^{(p)}e^{\lambda_1 z} - C_1^{(p)}e^{-\lambda_1 z}\right) + \right. \\ \left. + \frac{\lambda_2}{\left\|\alpha_2^{(p)}\right\|}\left(B_1^{(p)}e^{\lambda_2 z} - D_1^{(p)}\lambda_2 e^{-\lambda_2 z}\right)\right), \\ u_3^{(p)}(q,\omega,z) = -\frac{\lambda_1^2 + k_3^2}{\left\|\alpha_1^{(p)}\right\|}\left(A_1^{(p)}e^{\lambda_1 z} + C_1^{(p)}e^{-\lambda_1 z}\right) - \\ - \frac{\lambda_2^2 + k_3^2}{\left\|\alpha_2^{(p)}\right\|}\left(B_1^{(p)}e^{\lambda_2 z} + D_1^{(p)}e^{-\lambda_2 z}\right), \quad (10)$$

and norm of the vector is:

$$\left\|\alpha_n^{(p)}\right\| = \sqrt{q^2\lambda_n^2 c_1^2 + \left(\lambda_n^2 + k_3^2\right)^2},\ n = 1,\ 2\,. \quad (11)$$

In the relations, the quantities λ_n(8)-(10) are defined as the roots of such a biquadratic equation:

$$\lambda^4 + \left(k_1^2 + k_3^2 + c_1 c_3 q^2\right)\lambda^2 + k_1^2 k_3^2 = 0\,. \quad (12)$$

In contrast to the models used in previous papers, we will assume that the components ($u_1(z)$, $u_3(z)$) and components of the stress tensor (σ_{13} and σ_{33}) are continuous for the entire area of the nanosystem. This allows us to use the following boundary conditions:

$$\left[\begin{array}{l} u_{1(3)}^{(p)}(\omega,z)\Big|_{z=z_p} = u_{1(3)}^{(p+1)}(\omega,z)\Big|_{z=z_p} \\ \sigma_{13(33)}^{(p)}(\omega,z)\Big|_{z=z_p} = \sigma_{13(33)}^{(p+1)}(\omega,z)\Big|_{z=z_p}, \end{array}\right. . \tag{13}$$

where in the conditions (13):

$$\begin{array}{l} \sigma_{13}^{(p)}(\omega,z) = \dfrac{1}{2}C_{44}^{(p)}\left(-iqu_3^{(p)}(z) + \dfrac{du_1^{(p)}(z)}{dz}\right)e^{i(\omega t - qx)}, \\ \sigma_{33}(\omega,z) = \left(-iqC_{13}^{(p)}u_1^{(p)}(z) + C_{33}^{(p)}\dfrac{du_3^{(p)}(z)}{dz}\right)e^{i(\omega t - qx)}. \end{array} . \tag{14}$$

To determine the phonon spectrum, we proceed as follows. Let us take into account that in the environment, as applied to the nanosystem under study, there are only phonon waves incident on the nanostructure (to the left) and waves passing through it (to the right). This case is realized when the condition: $C_1^{(0)} = D_1^{(0)} = C_1^{(N+1)} = D_1^{(N+1)} = 0$ is satisfied.

Let us calculate the transmission coefficient for acoustic waves using the limit conditions and the transfer matrix method. Then the application of conditions (13) on all heteroboundaries gives the expression for transfer-matrix (4x4 matrix):

$$\begin{array}{l} T(q,\omega) = T^{(0,1)}(q,\omega)\times T^{(1,2)}(q,\omega)\times\ldots\times T^{(N,N+1)}(q,\omega) = \\ = \prod\limits_{i=1}^{N} T^{(i-1,i)}(q,\omega)\times T^{(i,i+1)}(q,\omega) \end{array} \tag{15}$$

Now, since:

$$\begin{array}{l} \left(A_1^{(0)} \quad 0 \quad B_1^{(0)} \quad 0\right)^T = T(q,\omega)\left(A_1^{(N+1)} \quad 0 \quad B_1^{(N+1)} \quad 0\right)^T, \\ T(q,\omega) = \begin{pmatrix} t_{11}^{(0,N+1)} & t_{12}^{(0,N+1)} & t_{13}^{(0,N+1)} & t_{14}^{(0,N+1)} \\ t_{21}^{(0,N+1)} & t_{22}^{(0,N+1)} & t_{23}^{(0,N+1)} & t_{24}^{(0,N+1)} \\ t_{31}^{(0,N+1)} & t_{32}^{(0,N+1)} & t_{33}^{(0,N+1)} & t_{34}^{(0,N+1)} \\ t_{41}^{(0,N+1)} & t_{42}^{(0,N+1)} & t_{43}^{(0,N+1)} & t_{44}^{(0,N+1)} \end{pmatrix}, \end{array} \tag{16))}$$

then the transmission coefficient is obtained by the following expression:

$$D(q,\omega) = \left|A_1^{(N+1)} / A_1^{(0)}\right|^2 = \left|B_1^{(N+1)} / B_1^{(0)}\right|^2. \tag{17}$$

The energies of the phonon spectrum are now obtained from the positions of the maxima for the function $D(q, \omega)$ ($\max D(q, \omega)$) and they are limited in the lower limit by the transverse phonon energies in the bulk GaN crystal, and in the upper limit by values of the order of 35 meV, which is the maximum energy for existing acoustic phonons.

Next, perform the calculation of group velocities using the following expression:

$$\upsilon_g(q) = d\omega_n / dq \, . \tag{18}$$

The desired value of thermal conductivity is determined according to the well-known relation [7]:

$$\begin{array}{l} \sigma_{ph}(q,T) = \dfrac{k_B^2 T}{3\hbar}\sum\limits_{n=1}^{N_{ph}}\int\limits_{x_{\min}}^{x_{\max}} \dfrac{x^2 e^x f_n(x)}{\left(e^x - 1\right)^2}\left[\upsilon_g(x)\right]^2 \tau_n(x)dx; \\ x_{\min} = \Omega_{\min} / kT; \; x_{\max} = \Omega_{\max} / kT, \end{array} \tag{19}$$

where the summation occurs over all phonon levels N_{ph}, k_B – is the Boltzmann constant, $\Omega_{\min}$ and $\Omega_{\max}$ determine the minimum and maximum values of the phonon spectrum energies respectively. The function that specifies the density of phonon states is defined as:

$$f_n(q,\omega) = \frac{q}{2\pi\upsilon_g(x)\sum\limits_{i=1}^{N}(z_i - z_{i-1})} \, . \tag{20}$$

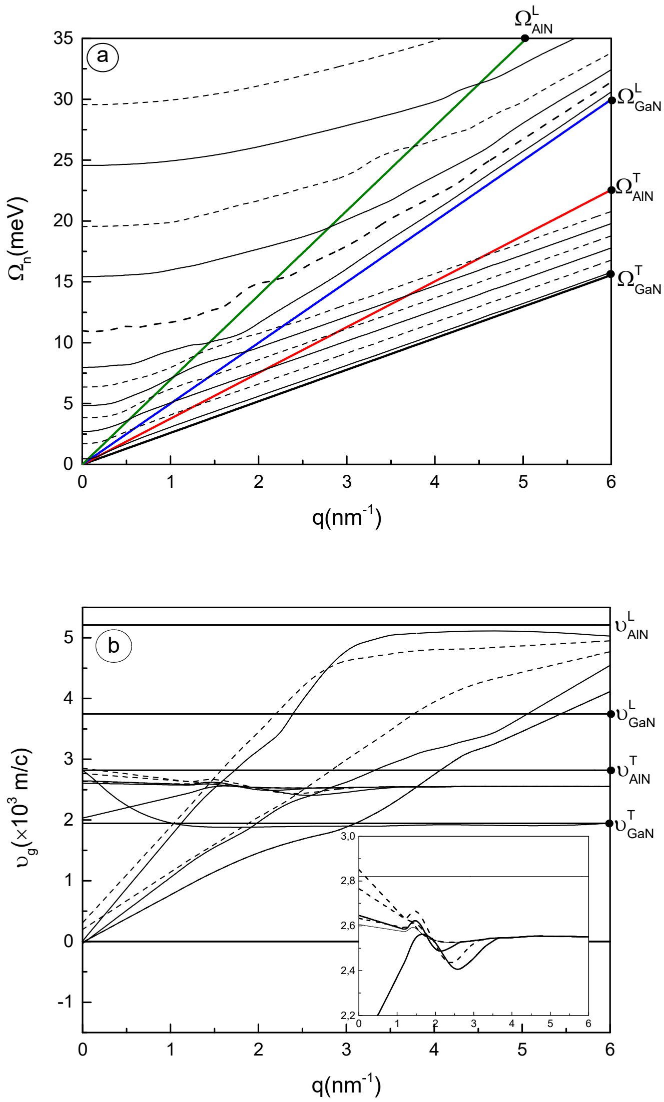

Fig. 2. Calculated dependences of the phonon spectrum and group velocities depending on the values of the wavevector q

The phonon relaxation time τ_n is defined as follows:

$$\tau_n(q) = \left(\tau_{ph}^{-1}(q) + \tau_U^{-1}(q)\right)^{-1}, \qquad (21)$$

with known constituents: $\tau_{ph}(q)$ – relaxation time on acoustic phonons, τ_U – relaxation time for Umklapp scattering [5, 8, 9].

III. Discussion of the results

We will use the developed theory to calculate the spectral characteristics of acoustic phonons and the thermal conductivity of the single cascade used in quantum cascade intersubband nanodevice, which was investigated experimentally in the paper [10]. The nanostructure under study contains 10 barrier layers made of AlN material, each 3 nm thick, and 9 potential well layers made of GaN material, each 4 nm thick.

In Fig. 2a, b are shown the calculated spectra Ω_n of acoustic phonons and the group velocities υ_g established according to the relation (17). It can be seen from Fig. 2a, that the energy spectrum of phonons consists of a large number of branches, demonstrating an almost linear dependence on the wave number q, except for a small interval in the vicinity of the origin, where their dependence has a quasi-square property, this is especially clearly seen for an increase in the energy values, which corresponds to this branches. It should be noted that it can be seen from Fig. 2b, the dependences of the group velocities for both branches are significantly different. So, for the lower group, they form in a narrow range, gradually merging into one curve. In this case, the dependences of the upper group change sequentially crossing the values of the group velocities for acoustic waves in the bulk. ($\upsilon_{\mathrm{GaN}}^{\mathrm{T}}, \upsilon_{\mathrm{AlN}}^{\mathrm{T}}$ - transverse waves, $\upsilon_{\mathrm{GaN}}^{\mathrm{L}}, \upsilon_{\mathrm{AlN}}^{\mathrm{L}}$ - longitudinal waves).

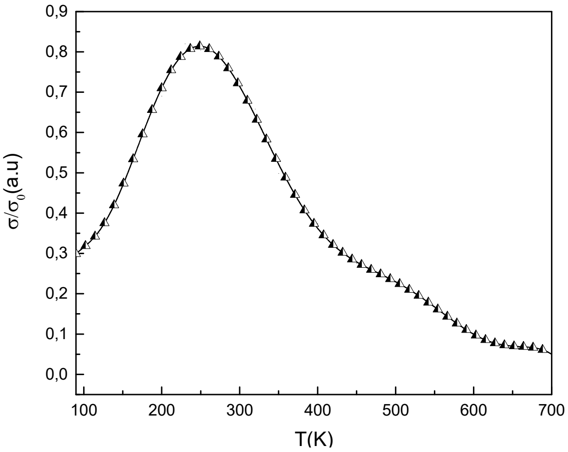

Fig. 3. Temperature dependence of the thermal conductivity of an investigated nanostructure

In Fig. 3 there are shown the temperature dependences of the thermal conductivity of the studied nanostructure calculated on the basis of the developed theory. For direct calculations, the relation (18) and the values of the phonon spectrum levels given in Fig. 2a were used. As can be seen from the figure, with increasing temperature, the conductivity first increases, forming a maximum in the range from 200K to 300K, and then gradually decreases. We note that the interval in which the maximum thermal conductivity is located actually corresponds to the temperature of operating mode for nanodevices based on nitride semiconductors.

If we compare the theoretical result obtained by us from the calculation of thermal conductivity with the result obtained in the experimental work [10], then we should conclude that there is a good correlation between these results. The theoretically obtained value of the maximum thermal conductivity of 252K is also in good agreement with the experimental value of 259K.

Conclusions

In the presented paper, an analytical theory of thermal conductivity of a nitride-based superlattice, a multilayer nanosystem, is developed and its spectrum is investigated in the case of acoustic phonons. On the basis of the developed theory, using the geometric and physical parameters of the experimentally studied nanosystem, direct calculations of the mentioned physical quantities were performed. It is established that the value of the maximum thermal conductivity is in the range from 200K to 300K with maximum localized at 252K. The results of the calculations will be useful to developers of electronic nanodevices operating in the high-frequency ranges of electromagnetic waves.

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