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THERMOELECTRIC FIGURE OF MERIT OF SEMICONDUCTOR SUPERLATTICES

The thermoelectric figure of merit of semiconductor superlattices has been studied in the quasi-classical one-miniband approximation. The change in the relaxation time of current carriers in 2D structures compared to their 3D analogs is taken into account when current carriers are scattered by acoustic phonons, point defects, and nonpolar optical phonons with arbitrary statistics. An analytical dependence of the figure of merit on the thermoelectric quality factor of the material and the width of the conduction miniband along the superlattice axis is established. It is shown that the figure of merit of semiconductor superlattices increases with increasing these parameters. Bibl. 14, Fig. 4.

Key words: superlattices, *conduction miniband,* relaxation time, thermopower, phonon thermal conductivity, thermoelectric figure of merit.

Introduction

Formulation of the problem. The efficiency of thermoelectric generators, as well as coefficient of performance of thermoelectric coolers is determined by the parameter of dimensionless thermoelectric figure of merit

$$zT = \frac{\alpha^2 \sigma}{\kappa_e + \kappa_{ph}} T, \qquad (1)$$

where α, σ, κ_e, κ_{ph} are the Seebeck coefficient, electric conductivity, electron and phonon thermal conductivity of thermoelectric material, T is average absolute temperature which characterizes the conditions of application of the thermoelectric device [1].

In [2], it is shown that for the bulk crystalline semiconductor materials, even with perfect combination of their parameters, $zT < 1.5$. In fact, the value of zT of modern thermoelectrics based on Bi_2Te_3 has approached the limit value of $zT < 1$.

Analysis of recent research and publications

In recent decades, intensive search for ways to increase the thermoelectric figure of merit has continued. In this case, the increase in thermoelectric figure of merit was mainly associated with a

decrease in phonon thermal conductivity while maintaining electrical conductivity at least at the level of degenerate wide-gap semiconductors. The use of low-dimensional semiconductor structures has been proposed: thin films [3, 4], superlattices, [5, 6], nanoscale structures [7]. Analysis of the experimental data confirms the promise of creating highly efficient thermoelectric materials based on nanotechnologies [8].

However, a decrease in the phonon component of the lattice thermal conductivity is not the only consequence of the influence of low-dimensional structures on the properties of a thermoelectric material: in nanostructured materials, the processes of transport and scattering of not only phonons, but also electrons can change [9, 10].

The purpose of the work is to study the thermoelectric figure of merit of semiconductor superlattices in quasi-classical one-miniband approximation with regard to a change in relaxation time in 2D structures as compared to their 3D analogs at scattering of current carriers by acoustic phonons, point defects and nonpolar optical phonons with arbitrary statistics.

Formulation of the task

Finding the distribution function

The physical properties of semiconductor materials are determined by their electronic spectrum, which has a strong anisotropy in superlattices. While the movement of current carriers in the direction perpendicular to the SL axis is almost free and corresponds to movement along a wide conduction band, the movement along the superlattice axis is limited.

In this direction, the electronic spectrum is miniband. In the case of sufficiently narrow layers, which are quantum wells for electrons, all electrons will be located near the bottom of the lower size quantization miniband. In the framework of quasi-classical approximation $2\varepsilon_0 >> \hbar / \tau_{2D}$, eE_0, $a\nabla_z k_0 T$ the law of electron dispersion in the lower miniband of SL is given by [11]

$$\varepsilon\left(\vec{k}\right) = \frac{\hbar^2 k_\perp^2}{2m_\perp} + 2\varepsilon_0\left(1 - \cos k_z a\right) \tag{3}$$

where $k_\perp = \left(k_x^2 + k_y^2\right)^{1/2}$, and k_z is transverse and longitudinal to SL axis components of quasi-wave vector, $m_\perp$ is transverse effective mass, which is close in size to the effective mass m^* of semiconductor electrons forming SL, a is SL period, $2\varepsilon_0$ is the width of SL conduction miniband in the direction k_z.

The calculation of kinetic coefficients will be carried out in the quasi-classical one-miniband approximation taking into account the change in the relaxation time in 2D structures in comparison with their three-dimensional 3D analogues. The nonequilibrium function of electron distribution f will be found from the kinetic Boltzmann equation

$$\vec{\upsilon}\frac{\partial f}{\partial \vec{r}} - e\vec{E}_0\frac{\partial f}{\hbar\partial \vec{k}} = -\frac{f_1}{\tau_{2D}}, \tag{2}$$

where $\vec{\upsilon} = \hbar^{-1}\partial\varepsilon\left(\vec{k}\right)/\partial\vec{k}$ is electron velocity, $\overrightarrow{E_0} = -\partial\varphi/\partial\vec{r}$ is electric field strength, φ is electric potential, $f_1 = f - f_0$, $f_0 = \left[1+\exp\left(\varepsilon-\varsigma\right)/k_0T\right]$ is equilibrium Fermi-Dirac distribution function with space-variable absolute temperature T and chemical potential ς, τ_{2D} is relaxation time, k_0 is Boltzmann constant.

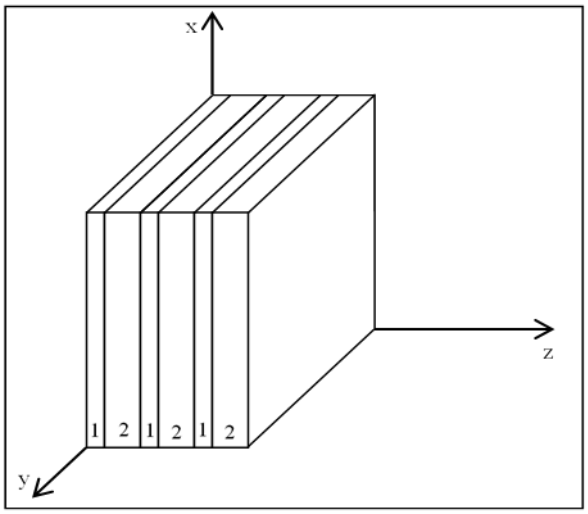

Fig.1 Schematic structure of semiconductor superlattice GaAs/AlAs. 1 – GaAs layer, 2 – AlAs layer.

The longitudinal component of the relaxation time tensor of the SL electron gas is written as follows [12]

$$\tau_{2D} = a\frac{2\sqrt{2m_{\perp}k_0T}}{3\pi\hbar}\tau_{3D}\left(\frac{\varepsilon}{k_0T}\right)^{1/2}, \tag{3}$$

where

$$\tau_{3D} = \tau_0\left(\frac{\varepsilon}{k_0T}\right)^{r-1/2}, \tag{4}$$

– relaxation time in the bulk sample, τ_0 – electron energy-independent constant, r – scattering parameter are given by:

on point defects (short-term potential)

$$\tau_0 = \frac{\pi\hbar^4}{m_n\left(2m_nk_0T\right)^{1/2}U_0^2N_g}, \tag{5}$$

on acoustic phonons

$$\tau_0 = \frac{2\pi\hbar^4\rho\upsilon_0^2}{E_1^2\left(2m_nk_0T\right)^{3/2}}, \tag{6}$$

on nonpolar optical phonons ($k_0T >> \hbar\omega_0$)

$$\tau_0 = \frac{2}{\pi}\left(\frac{\hbar\omega_0}{E_0}\right)^2 \frac{\hbar^2 a^2 \rho}{m_n (2m_n k_0 T)^{3/2}}, \tag{7}$$

It is obvious that the differences in the power dependence of the relaxation time of the bulk sample and the superlattice are due to the different energy dependence of the density of states.

Solving equation (2) in the relaxation time approximation, for the nonequilibrium additive to the Fermi-Dirac distribution function we obtain

$$f_1 = -\tau_{2D}\left(\frac{\partial f_0}{\partial \varepsilon}\right)\left[\frac{\varepsilon(\vec{k})-\varsigma}{T}\ \vec{\upsilon}\nabla T - e\vec{\upsilon}\nabla\left(\varphi - \frac{\varsigma}{e}\right)\right]. \tag{8}$$

It is obvious that the first component of the nonequilibrium additive is caused by the sample temperature gradient, and the second one is caused by the current carrier energy gradient.

Calculation of kinetic coefficients

The current density and energy flux density are found from the known relations [12]

$$\vec{j} = -\frac{2e}{(2\pi)^3}\int \vec{\upsilon}(\vec{k}) f_1(\vec{k},\vec{r})\ d\vec{k}, \tag{9}$$

$$\vec{w} = \frac{2}{(2\pi)^3}\int \left[\varepsilon(\vec{k})-\varsigma\right] \vec{\upsilon}(\vec{k}) f_1(\vec{k},\vec{r})\ d\vec{k} \tag{10}$$

We assume that the vectors $\overrightarrow{E_0}$ and ∇T are directed along the superlattice axis, which is compatible with the axis of the cylindrical coordinate system *0z*. Given (8) and integrating in the cylindrical coordinate system for current and energy density, we obtain

$$j = j_z = \sigma(\eta,\beta)\ \nabla_z\left(\frac{\varsigma}{e} - \varphi\right) - \alpha(\eta,\beta)\ \sigma(\eta,\beta)\ \nabla_z T \tag{11}$$

$$w = w_z = -\kappa_e(\eta,\beta)\ \nabla_z T \tag{12}$$

where $\alpha(\eta, \beta)$, $\tau(\eta, \beta)$, $\kappa_e(\eta, \beta)$ are the Seebeck coefficient, electric conductivity and electron component of thermal conductivity along the SL axis.

Taking into account the form of the distribution function, from (11) and (12) we find the kinetic coefficients.

The Seebeck coefficient

$$\alpha(\eta,\beta)=\frac{\nabla_z\left(\frac{\varsigma}{e}-\varphi\right)}{\nabla_z T}=-\frac{k_0}{e}\left[\frac{I_{1,2,0}(\eta,\beta)+\beta\ I_{0,2,2}(\eta,\beta)}{I_{0,2,0}(\eta,\beta)}-\eta\right], \tag{13}$$

electric conductivity

$$\sigma(\eta,\beta)=\sigma_0\beta^2 I_{0,2,0}(\eta,\beta), \tag{14}$$

electron component of thermal conductivity

$$\kappa_e(\eta,\beta)=L_{2D}(\eta,\beta)\ \sigma(\eta,\beta)T, \tag{15}$$

where

$$L_{2D}(\eta,\beta)=\left(\frac{k_0}{e}\right)^2\left\{\begin{array}{l}\dfrac{I_{2,2,0}(\eta,\beta)+2\beta\ I_{1,2,2}(\eta,\beta)+\beta^2 I_{0,2,4}(\eta,\beta)}{I_{0,2,0}(\eta,\beta)}-\\ -\left[\dfrac{I_{1,2,0}(\eta,\beta)}{I_{0,2,0}(\eta,\beta)}+\beta\dfrac{I_{0,2,2}(\eta,\beta)}{I_{0,2,0}(\eta,\beta)}\right]^2\end{array}\right\} \tag{16}$$

the Lorentz number,

$$I_{k,l,m}(\eta,\beta)=\int_0^{\pi}F_k(\eta,z,\beta)(\sin z)^l\left(\sin\frac{z}{2}\right)^m dz, \tag{17}$$

$$F_k(\eta,z,\beta)=\int_0^{\infty}\frac{\exp\left(x-\eta+\beta\ \sin^2\frac{z}{2}\right)}{\left[1+\exp\left(x-\eta+\beta\ \sin^2\frac{z}{2}\right)\right]^2}x^k dx, \tag{18}$$

three-parameter Fermi integrals [12],
$\varepsilon_\perp=\hbar^2k_\perp^2/2m_\perp$, $x=\varepsilon_\perp/k_0T$, $\eta=\varsigma/k_0T$, $z=ak_z$, $\beta=2\varepsilon_0/k_0T$ is reduced width of conduction miniband in the direction of superlattice axis.

Calculation of thermoelectric figure of merit

Using the found kinetic coefficients, we calculate the thermoelectric figure of merit of *2D*-structure

$$z_{2D}(\eta,\beta)T=\frac{\alpha^2(\eta,\beta)\ \sigma(\eta,\beta)}{\kappa_e(\eta,\beta)+\kappa_{ph}}T=\frac{\alpha_0^2(\eta,\beta)}{L_0(\eta,\beta)+B^{-1}\left[\beta^2 I_{0,2,0}(\eta,\beta)\right]^{-1}}, \tag{19}$$

where $L_0(\eta,\beta) = L_{2D}(\eta,\beta)/(k_0/e)^2$ is the reduced Lorentz number, $\alpha_0(\eta,\beta) = \alpha(\eta,\beta)/(-k_0/e)$ is the reduced Seebeck coefficient,

$B = \left(\frac{k_0}{e}\right)^2 \frac{\sigma_0 T}{\kappa_{ph}}$ is coefficient of thermoelectric quality which contains material parameters of 2D lattice: coefficient of phonon thermal conductivity, SL period, effective mass and mobility of current carriers in the direction of SL axis and has a significant impact on the value of thermoelectric figure of merit. A similar parameter was introduced for the bulk semiconductor samples in [2].

Analysis of the results

At fixed values of current carrier parameters *B* and β, the thermoelectric figure of merit is a function of only η, and the dependence *z(*η*)* has an extremum. That is, there is an optimal concentration of doping impurities, which leads to maximum figure of merit values. After maximization of *z(*η*)* by chemical potential, the effect of parameters *B* and β on the thermoelectric figure of merit was investigated. Estimation of the most realistic range of changes in thermoelectric quality factor gives *B = 0.075-0.125.*

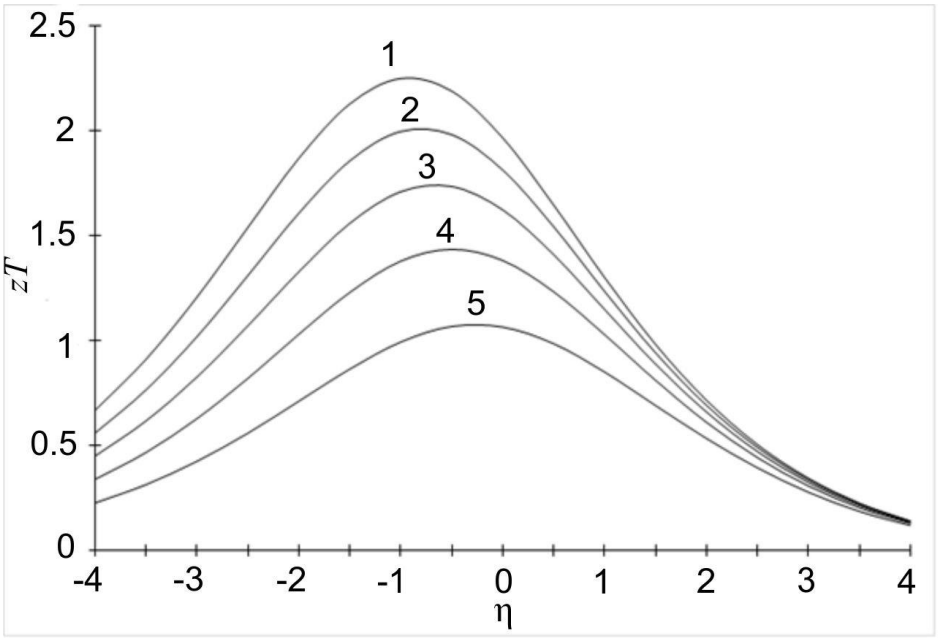

Fig. 2 Dependence of thermoelectric figure of merit on the reduced chemical potential at the width of the conduction miniband β=5 and at different values of quality factor. Curve 1 at B =0.15, 2- B =0.125, 3 - B =0.1, 4 - B =0.075, 5 - B =0.05.

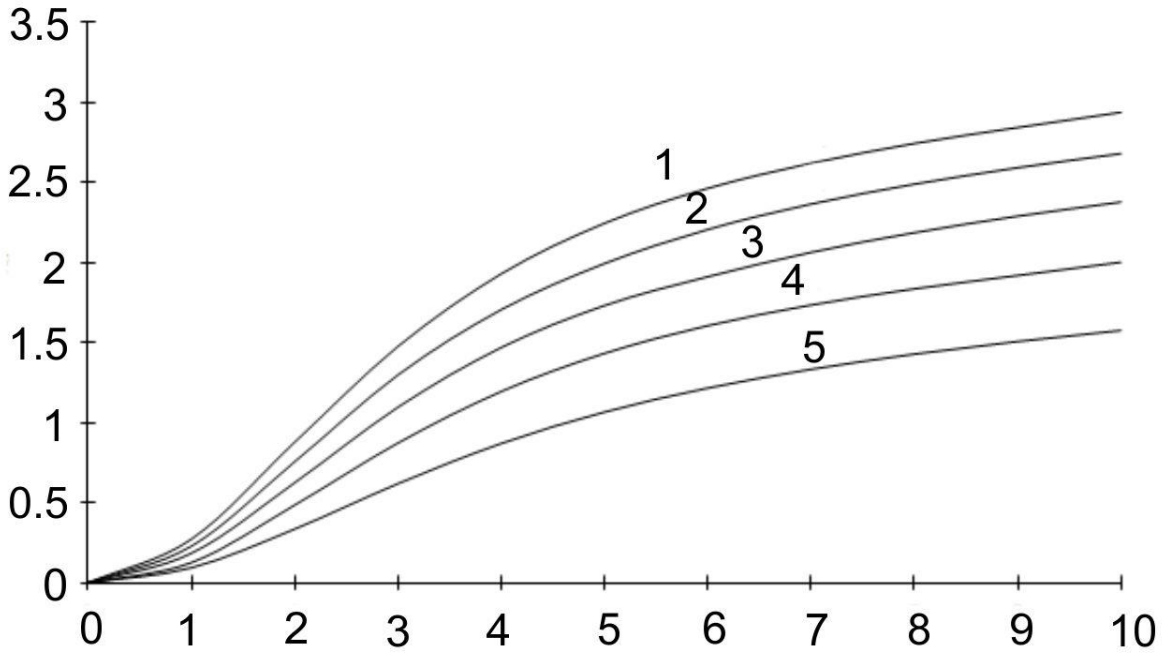

Fig. 3. Dependence of maximum thermoelectric figure of merit on the width of conduction miniband at different values of quality factor. Curve 1 at B =0.15, 2- at B =0.125, 3 – at B =0.1, 4 at - B =0.075, 5 at- B =0.05.

Taking into account the finiteness of the width of the conduction miniband leads to the dependence of the Lorentz number on its width. In particular, as the width of the miniband decreases in the direction of the SL axis, the Lorentz number and the electrical conductivity and electronic component of thermal conductivity decrease. And, despite the decrease in phonon thermal conductivity, this process leads to an overall decrease in thermoelectric figure of merit.

On the contrary, increasing the width of the conduction miniband increases the conductivity. However, this increases the electronic component of thermal conductivity, which can compensate for the decrease in phonon thermal conductivity. This process can lead to an overall reduction in thermoelectric figure of merit. It is obvious that there is an optimal width of the conduction miniband, which for the GaAs /AlAs superlattices is in the range $\beta = 5\text{-}9$.

In two-dimensional structures for the statistics of degenerate gas of current carriers, the reduced Lorentz number reaches the maximum value $L_0(\infty, \beta)=\pi^2/3$, and for nondegenerate electron gas and ultra-narrow minimum band of minimum conductivity - $L_0(\eta, 0)=1$.

To verify the correctness of the obtained relations, a boundary transition was performed at $\beta \to \infty$. The increase in β should be considered as the approximation of the width of the conduction band in the direction parallel to the SL axis to the width of the wide conduction band in the direction $k_\perp$.

In the boundary case $\beta = \infty$, the analytical relations for the kinetic coefficients are transferred to the known formulae for the bulk semiconductors with a parabolic dispersion law [12].

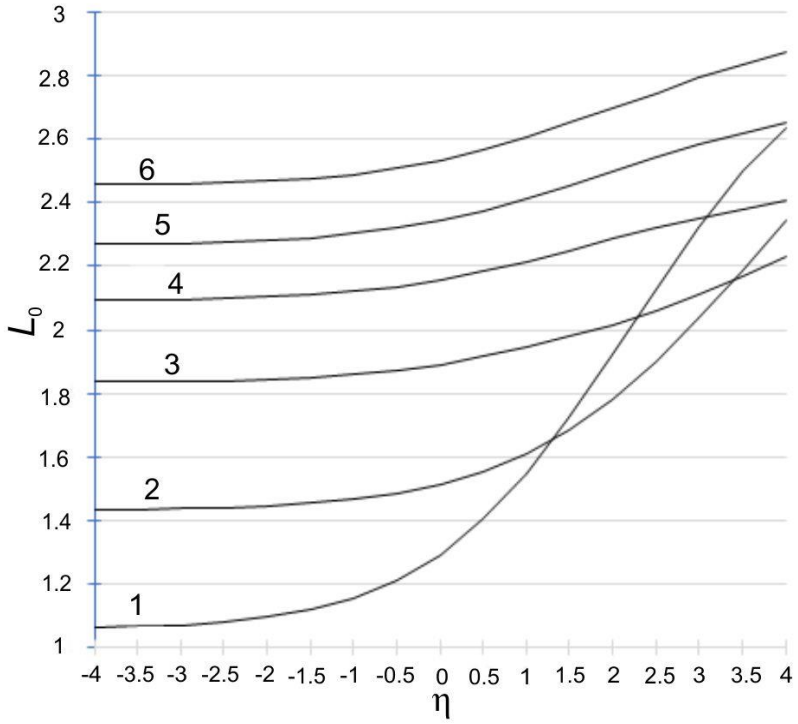

Fig. 4. Dependence of reduced Lorentz number on chemical potential at different width of conduction miniband. Curves: 1 - at β=1, 2 – at β =3, 3 at β =5, 4 at β =7, 5 at β =10, 6 - at β =∞.

One of the most important results of the creation of low-dimensional heterostructures, leading to an increase in thermoelectric figure of merit, is a decrease in the thermal conductivity of the lattice as a result of phonon scattering on surfaces and heterointerfaces.

However, the decrease in lattice thermal conductivity is not the only consequence of the impact of low-dimensional structures on the properties of thermoelectric material: in nanostructured materials, the scattering processes of not only phonons but also of electrons change. In particular, the relaxation time of current carriers in 2D structures changes in comparison with their 3D analogues.

Conclusions

The thermoelectric figure of merit of superlattices depends on the chemical potential η, the width of conduction miniband β and thermoelectric quality factor *B*. The dimensionless parameter of thermoelectric material quality is determined by the value: phonon thermal conductivity, superlattice period, effective mass of current carriers in the direction of superlattice axis, etc. The most realistic range of change in *B* for two-dimensional structures based on GaAs/AlAs is *0.050-0.125*.

At fixed values of parameters *B* and β, the thermoelectric figure of merit is a function of only η, and the dependence *z(η)* has an extremum which, when *B* increases, shifts toward smaller values of chemical potential. In the region of room temperatures, under the condition of the optimal

concentration of dopants, with the width of the conduction miniband $\beta=10$ and with the most favorable value of thermoelectric quality factor $B=0.125$, the thermoelectric figure of merit of superlattices based on *AlGaAs* can reach the values $zT=3$. The resulting limit value of SL thermoelectric figure of merit is sufficient for thermoelectric devices to compete in efficiency with electric generators and refrigerators operating on other principles.

Naturally, the effect of electron-phonon drag can make significant adjustments to the value of thermoelectric figure of merit [13, 14].

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ТЕРМОЕЛЕКТРИЧНА ДОБРОТНІСТЬ НАПІВПРОВІДНИКОВИХ НАДҐРАТОК

В квазікласичному одномінізонному наближені досліджена термоелектрична добротність напівпровідникових надґраток. Врахована зміна часу релаксації носіїв струму 2D структурах в порівнянні з їх 3D аналогами при розсіюванні носіїв струму на акустичних фононах, точкових дефектах і неполярних оптичних фононах при довільній статистиці. Встановлена аналітична залежність добротності від коефіцієнта термоелектричної якості матеріалу та ширини мінізони провідності в напрямку осі надгратки. Показано, що добротність напівпровідникових надґраток збільшується із збільшенням цих параметрів. Бібл. 14, рис. 4
Ключові слова: надгратки, *мінізона провідності,* час релаксації, термоЕРС, фононна теплопровідність, термоелектрична добротність.

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ТЕРМОЭЛЕКТРИЧЕСКАЯ ДОБРОТНОСТЬ ПОЛУПРОВОДНИКОВЫХ СВЕРХРЕШЕТОК

В квазиклассическом одноминизонном приближении исследована термоэлектрическая добротность полупроводниковых сверхрешеток. Учтено изменение времени релаксации носителей тока в 2D структурах по сравнению с их 3D аналогами при рассеивании носителей тока на акустических фононах, точечных дефектах и неполярных оптических фононах при произвольной статистике. Установлена аналитическая зависимость добротности от коэффициента термоэлектрического качества материала и ширины минизоны проводимости по оси сверхрешётки. Показано, что добротность полупроводниковых сверхрешеток увеличивается с увеличением этих параметров. Библ. 14, рис. 4.

Ключевые слова: сверхрешетки, минизона проводимости, время релаксации, термоЭДС, фононная теплопроводность, термоэлектрическая добротность, .

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