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Vladislav S. Volcheck, Maryia S. Baranova, Viktor R. Stempitsky

Belarusian State University of Informatics and Radioelectronics, Minsk, Republic of Belarus

THERMAL CONDUCTIVITY OF WURTZITE GALLIUM NITRIDE

Abstract. This paper reviews the theoretical and experimental works concerning one of the most important parameters of wurtzite gallium nitride – thermal conductivity. Since the heat in gallium nitride is transported almost exclusively by phonons, its thermal conductivity has a temperature behavior typical of most nonmetallic crystals: the thermal conductivity increases proportionally to the third power of temperature at lower temperatures, reaches its maximum at approximately 1/20 of the Debye temperature and decreases proportionally to temperature at higher temperatures. It is shown that the thermal conductivity of gallium nitride (depending on fabrication process, crystallographic direction, concentration of impurity and other defects, isotopic purity) varies significantly, emphasizing the importance of determining this parameter for the samples that closely resemble those being used in specific applications. For isotopically pure undoped wurtzite gallium nitride, the thermal conductivity at room temperature has been estimated as high as 5.4 W/(cm·K). The maximum room temperature value measured for bulk-shaped samples of single crystal gallium nitride has been 2.79 W/(cm·K).

Keywords: gallium nitride, phonon, thermal conductivity, GaN, temperature dependence

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В. С. Волчэк, М. С. Баранова, В. Р. Стемпицкий

Белорусский государственный университет информатики и радиоэлектроники, Минск, Республика Беларусь

ТЕПЛОПРОВОДНОСТЬ НИТРИДА ГАЛЛИЯ С КРИСТАЛЛИЧЕСКОЙ СТРУКТУРОЙ ТИПА ВЮРЦИТА

Аннотация. Выполнен анализ теоретических и экспериментальных исследований одного из важнейших параметров нитрида галлия с кристаллической структурой типа вюрцита – теплопроводности. Так как перенос тепла в нитриде галлия осуществляется главным образом с помощью фононов, его теплопроводность имеет температурную зависимость, характерную для большинства неметаллических кристаллов: увеличивается пропорционально третьей степени температуры в области низких температур, достигает своего максимального значения при температуре, приблизительно равной 1/20 от дебаевской, и уменьшается пропорционально температуре в области высоких температур. Показано, что в зависимости от условий (технология изготовления образца, кристаллографическое направление, концентрация примеси и других дефектов, изотопный состав) теплопроводность нитрида галлия может находиться в большом диапазоне значений, что указывает на важность определения этого параметра именно тех образцов материала, которые используются в конкретных приложениях. Теплопроводность нелегированного изотопно-чистого нитрида галлия при комнатной температуре оценивается на уровне 5,4 Вт/(см·К). Максимальная теплопроводность, достигнутая для объемного образца из монокристаллического нитрида галлия, равна 2,79 Вт/(см·К).

Ключевые слова: нитрид галлия, теплопроводность, фонон, GaN, температурная зависимость

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Introduction. Over the last decades, a lot of attention has been focused on gallium nitride (GaN) as a promising material for the application in high-power radio-frequency electronic and optoelectronic devices [1, 2]. The intense interest in this semiconductor arises from the following factors. First, the wide band gap of GaN yields a high breakdown voltage, which is valuable for high-power device operation. Second, the saturation velocity of electrons in GaN is over a factor of two larger than that in silicon, which enables high-frequency operation. Furthermore, the natural structure of GaN is wurtzite – a hexagonal crystal structure where electrical polarization effects play a major role [3]. At heterojunctions formed by group III nitrides the polarization induces a very high electron concentration, providing a large current density.

Although the structural, electronic and optical properties have been extensively studied, relatively little work, both analytical and experimental, has to date been reported on the thermal conductivity κ of GaN. Meanwhile, this thermoelectric parameter – a measure of the ability to conduct heat – is significant from both fundamental and applied aspects. One of the critical issues in the development of the nitride high-power technology is self-heating. A non-uniform distribution of the dissipated power and a rise of the average temperature that lead to the formation of a hot spot near the active area and result in the degradation of the drain current and output power are characteristic of GaN high electron mobility transistor – a device considered for broadband/multiband communication applications, radar systems and electronics in harsh environments [4, 5]. For this reason, the monitoring of the average and maximum temperatures inside the device structure is crucial to its efficiency and reliability. Being an important part of technology computer-aided design, thermal simulation makes it possible for device engineers to identify the principal cause of thermal problems as well as to create, optimize and verify the thermal design. The simulation of self-heating implies the addition of a lattice heat flow equation to the set of the fundamental semiconductor device equations – the Poisson equation and the continuity equations. Standard device simulators implement a thermodynamically rigorous model of lattice heating that accounts for Joule heating, heating and cooling due to carrier generation and recombination, the Peltier and Thomson effects [6]. One of the input parameters of the lattice heat flow equation is thermal conductivity, which can depend on temperature T and other factors. This is critical to take into account in thermal simulation as the result can be highly sensitive to the thermal conductivity values for certain regions of the device structure. Otherwise, a significant error may occur. Due to the recent advancements in the ammonothermal and hydride vapor phase epitaxy (HVPE) methods, free-standing GaN substrates have begun to gain in popularity lately. The use of native electrically insulating substrates enables to improve the crystalline quality of epitaxial layers by preventing the necessity of growing a nucleation layer, which would act as a thermal barrier for the heat flow generated in the active area of the device structure [7, 8]. As opposed to other common substrate materials, particularly sapphire, GaN may provide a sufficient heat dissipation efficiency without recourse to additional heat-removing elements [9]. From this aspect, the thermal conductivity of GaN is of interest as a key design parameter.

In this paper, we present a review of the works, both theoretical and experimental, concerning the thermal conductivity of wurtzite GaN, mainly its dependence on temperature. The problem of determining the value of this parameter has a long history and has been tackled by many authors. The correlation and interpretation of the published data is quite challenging as the densities of impurity atoms, vacancies, dislocations and isotopes in various samples are often very different or even unknown. The theoretical limit at a temperature of 300 K for isotopically pure undoped wurtzite GaN is estimated as high as 5.4 W/(cm·K) [10]. However, values lower than 3.0 W/(cm·K) are prevalent for bulk samples grown on foreign substrates owing to the presence of imperfections in the crystal [11].

Thermal Conduction in Nonmetallic Crystals. The internal energy may be transferred within a solid by several independent agents. In metals, the heat is carried both by electrons and by lattice waves, or phonons, although the electronic contribution is predominant. On the other part, in insulators, the heat is carried almost entirely by phonons. From a thermal conduction point of view, semiconductors are very similar to insulators, with the compounds listed above being no exceptions. Thus, the transport of heat in nonmetallic crystals can be interpreted using the concept of a phonon [12].

The atoms of a crystal are not quite stationary and take part in thermal vibrations around their equilibrium positions. Instead of describing the individual vibrations of the particles, which is unrealistic

and impractical, their collective motion in the crystal is considered. This approach is based on the fact, that powerful interatomic forces immediately transmit the vibrations of one particle to other particles and a collective motion in the form of an elastic wave involving all the particles of the crystal is excited in it. Such elastic waves are termed normal modes. The quantity of the normal modes equals to the number of degrees of freedom, which is $3N$, where N is the number of particles constituting the crystal. The frequency ω and the wavelength λ of a normal mode are linked by

$$\omega = \frac{2\pi\nu}{\lambda}, \quad (1)$$

where ν is the velocity of wave propagation (of sound).

Wave processes are usually described by a wave-vector \vec{q} , whose direction coincides with that of wave propagation and whose absolute value is

$$q = \frac{2\pi}{\lambda}. \quad (2)$$

The relationship between ω and q is termed the dispersion relationship, which is given by

$$\omega = \nu q. \quad (3)$$

The lattice waves in a crystal occupy some spectrum of frequencies. The temperature at which the entire spectrum, including the mode with the maximum frequency, is excited is called the Debye temperature θ_D . A rise in temperature above θ_D shall not be accompanied by the appearance of new normal modes. In this case, the role of the temperature is to increase the intensity, or the energy, of each of the normal modes. According to the oscillation theory, the energy of a normal mode equals to the energy of an oscillator with a mass equivalent to the mass of the vibrating atoms and the frequency of the normal mode. Hence, the total energy of the crystal formed by N atoms taking part in coupled vibrations equals to the energy of $3N$ independent normal harmonic linear oscillators. The energy of a quantum oscillator is written as

$$E = \left(n + \frac{1}{2} \right) \hbar\omega, \quad (4)$$

where n is a quantum number and \hbar is the reduced Planck constant.

The minimum portion of energy that can be emitted or absorbed by a lattice in the process of thermal vibrations corresponds to the transition of the normal mode being excited from the given energy level to the adjacent level and equals to $\hbar\omega$. This portion, or quantum, of energy is termed a phonon.

The vibrations of the atoms in a crystal can only be treated as the normal modes of the lattice under certain idealized conditions: the interatomic forces are harmonic (obey Hooke's law) and the lattice is structurally perfect, isotopically pure and has no external boundaries. The departure from these idealized conditions leads to an anharmonic nature of the atomic vibrations. There are two phenomena caused by the anharmonicity – thermal expansion and thermal resistance. The effect that the thermal expansion of a lattice has on its vibrational properties is described by the Grüneisen constant γ . This dimensionless parameter relates the thermal expansion coefficient to the heat capacity defined by harmonic contribution:

$$\gamma = \frac{B\alpha_V}{C_V}, \quad (5)$$

where B is the bulk modulus, α_V is the volumetric thermal expansion coefficient and C_V is the volumetric heat capacity.

Should the vibrations of the atoms be strictly harmonic propagating through a lattice in the form of noninteracting elastic waves, there could be no thermal resistance in the crystal. In the absence of interaction the waves would be able to spread without scattering – that is, without encountering any resistance – and the thermal conductivity of the crystal would be infinitely high. The anharmonicity deprives the normal modes of the lattice of their independence and forces them to interact, exchanging energy and changing the direction of their propagation through mutual scattering. It is just these processes of interaction between the elastic waves that enable the transfer of energy from the modes of one frequency to the modes of another and the establishment of thermal equilibrium in the crystal.

It is well known that the thermal conductivity of silver at room temperature is $4.3 \text{ W}/(\text{cm} \cdot \text{K})$ – highest of any metal. For copper, $\kappa = 4.0 \text{ W}/(\text{cm} \cdot \text{K})$. There are several nonmetallic crystals that exhibit a high thermal conductivity comparable to or even exceeding that of silver and copper. Many of these crystals are adamantine, or diamond-like, compounds, such as diamond, boron nitride, silicon carbide, beryllium oxide, boron phosphide, aluminum nitride, beryllium sulphide, boron arsenide, silicon, aluminum phosphide and gallium phosphide. GaN is also included in this category of materials. In all of these crystals the atoms possess tetrahedral coordination and κ in this series decreases from $20 \text{ W}/(\text{cm} \cdot \text{K})$ for diamond to $1 \text{ W}/(\text{cm} \cdot \text{K})$ for gallium phosphide [13].

The dependence of the thermal conductivity on temperature is universal for almost all pure electrically insulating crystals: κ grows as T^3 at lower temperatures, reaches a maximum at approximately $0.05\theta_D$ and falls off as T^{-1} or faster at higher temperatures [14]. At temperatures below θ_D , the phonon concentration reduces dramatically with a fall in temperature. This leads to a sharp increase in the phonon mean free path – the average distance between scattering events, or collisions – so that at $0.05\theta_D$ it becomes comparable to the dimensions of the crystal. As the crystal surface is usually a poor reflector of phonons, any further reduction in temperature does not result in an increase in the mean free path, for the latter is determined by the dimensions of the crystal only. The temperature dependence of the thermal conductivity within this range of temperatures parallels that of the heat capacity, which is proportional to T^3 .

As the temperature rises, there are two opposing processes occurring at the same time – an increase in the phonon concentration, which should make the thermal conductivity to grow, and an increase in the resistive phonon-phonon scattering – three-phonon umklapp scattering or U-process – and a subsequent decrease in the phonon mean free path, which should cause the thermal conductivity to drop. For a low phonon concentration, the first factor is predominant and κ increases with temperature. However, starting with a certain concentration of phonons the second factor becomes prevailing and κ after passing through its maximum decreases with temperature as T^{-1} or faster.

Phonons can also be scattered by point defects, such as impurity atoms, vacancies and isotopes. The reasons for this are the differences in mass, size and interatomic force constants between the defects and the host atoms. Having no effect at low temperatures, point defects, however, take on significance at intermediate temperatures, where they will depress the peak of the thermal conductivity below that of the pure crystal. The magnitude of an isotope effect can be determined by

$$P = \frac{\kappa_{\text{pure}}}{\kappa_{\text{natural}}}, \quad (6)$$

where κ_{pure} corresponds to κ of the isotopically pure material and κ_{natural} to κ with naturally occurring isotope proportions.

While the influence of most scattering processes on overall thermal conductivity at different temperatures has been well analyzed, the normal three-phonon scattering processes, or N-processes, have been neglected in many publications. This scattering mechanism conserves phonon momentum and energy and hence does not contribute directly to thermal resistance. However, the normal processes lead to a redistribution of momentum and energy among phonons more likely to participate in umklapp processes thereby indirectly affecting the overall thermal conductivity.

In many nonmetallic crystals, the electronic component of the thermal conduction is small enough and can be neglected. Florescu *et al.* [15] estimated the electronic contribution to the thermal conductivity of GaN to be about $1.5 \cdot 10^{-3} \text{ W}/(\text{cm} \cdot \text{K})$, which is three orders of magnitude smaller than the typical values of κ for GaN.

Theoretical Studies of the Thermal Conduction in GaN. One of the earliest theoretical studies of the thermal conduction in GaN was carried out by Slack [13], who applied the fundamental theory given by Leibfried and Schlömann to predict the intrinsic thermal conductivity – the upper limit for κ defined only by crystal anharmonicity. According to this theory, the thermal conductivity of adamantine crystals in a temperature range of $T > \theta_D$ is proportional to the factor $\delta m \theta_D^3$:

$$\kappa = A \frac{\delta m \theta_D^3}{\gamma^2 T}, \quad (7)$$

where A is a scaling constant, δ^3 is the average volume occupied by one atom of the crystal and m is the average mass of the atoms in one mole of the compound.

It must be noted that Eq. (7) is strictly valid only for face-centered-cubic crystals having one atom per primitive crystallographic unit cell. From the dependencies of κ on $\delta m \theta_D^3$ plotted for the series of materials, for which the data was by then available, the scaling constant was estimated at $2.5 \text{ s}^{-3} \cdot \text{K}^{-3}$. For GaN, $\delta = 2.25 \cdot 10^{-8} \text{ cm}$, $m = 41.86 \text{ g}$, $\theta_D = 600 \text{ K}$ and $\delta m \theta_D^3 = 203.44 \text{ cm} \cdot \text{g} \cdot \text{K}^3$. The Grüneisen parameter was omitted as it was yet not known. Thus, the intrinsic thermal conductivity at 300 K was predicted at $1.7 \text{ W}/(\text{cm} \cdot \text{K})$.

A similar approach but without using a scaling constant was taken by Witek [16], who formulated the thermal conductivity of ideal single nonmetallic crystals in terms of the bulk modulus, the Debye temperature and the Grüneisen parameter:

$$\kappa = \frac{1}{3} \frac{\delta \nu B}{\gamma^2 T}. \tag{8}$$

This equation was derived from the suggestion by Dugdale and MacDonald [17] that the thermal conductivity should be related to the thermal expansion coefficient. They noticed that the dimensionless term $\gamma \alpha_V T$ serves as a measure of the departure of a lattice from harmonicity and proposed that the phonon mean free path could be calculated by

$$l = \frac{\delta}{\gamma \alpha_V T}. \tag{9}$$

The substitution of B in Eq. (8) by $\frac{\gamma C_V}{\alpha_V}$ and the insertion of Eq. (9) give another expression for thermal conductivity:

$$\kappa = \frac{1}{3} l \nu C_V. \tag{10}$$

By solving Eq. (8), the Grüneisen parameter and the intrinsic thermal conductivity of GaN at 300 K were estimated at 0.74 and $4.1 \text{ W}/(\text{cm} \cdot \text{K})$, respectively. The latter exceeded considerably any values that had previously been obtained by experiment. In real lattices, the anharmonicity is not the only source of the resistive phonon scattering. Macroscopic and point defects scatter phonons independently on the anharmonicity, decreasing thermal conductivity.

The evaluation of the thermal conductivity of a GaN sample characterized by realistic material parameters specific for a given growth technique can be made in the framework of Callaway’s phenomenological model [18]. It has proved to be a feasible computational tool and has been widely used for technologically important semiconductors. A long list of the publications in which this model was employed is presented in [19].

In Callaway’s formulation, the thermal conductivity contains two terms $\kappa = \kappa_1 + \kappa_2$, where

$$\kappa_1 = \frac{\kappa_B^4 T^3}{2\pi^2 \hbar^3 \nu} \int_0^{\theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau dx, \tag{11}$$

$$\kappa_2 = \frac{\kappa_B^4 T^3}{2\pi^2 \hbar^3 \nu} \frac{\left\{ \int_0^{\theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \frac{\tau}{\tau_N} dx \right\}^2}{\int_0^{\theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \frac{\tau}{\tau_N \tau_R} dx}, \tag{12}$$

where κ_B is the Boltzmann constant, τ is the combined relaxation time, τ_N and τ_R are the relaxation times in normal and resistive processes, respectively, and

$$x = \frac{\hbar \omega}{\kappa_B T}. \tag{13}$$

This model is derived using Debye-like phonon density of states and under the assumption of one effective acoustic-phonon dispersion branch and the additivity of the relaxation rates for independent scattering mechanisms:

$$\frac{1}{\tau} = \frac{1}{\tau_N} + \frac{1}{\tau_R}. \quad (14)$$

The relaxation rates in normal processes become comparable to those in resistive processes only in high-purity, defect-free samples. Otherwise, the resistive processes are totally dominant and the condition $\tau_N \gg \tau_R$ leads to $\kappa \approx \kappa_1$. Thus, the final expression is often written as

$$\kappa = \frac{\kappa_B^4 T^3}{2\pi^2 \hbar^3 v} \int_0^{\theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau dx \approx \frac{1}{3} \sum_{j,q} v_j^2(\vec{q}) \tau_j(\vec{q}) C_{V_j}(\vec{q}), \quad (15)$$

where j denotes a specific phonon polarization type.

Using Callaway's theory, the thermal conduction in GaN has been investigated by a number of researchers [10, 14, 20, 21]. Kotchetkov *et al.* [20] considered the three-phonon umklapp scattering (described by a relaxation time τ_U), the scattering on impurity atoms and the scattering on core, screw and edge dislocations. Given the wide discrepancy in the material parameters reported for GaN, the calculations were performed with two distinctly different sets of the input data. The solution of Eq. (15) with $\tau = \tau_U$ gave the values of 3.44 and 3.70 W/(cm · K) for the intrinsic thermal conductivity at 300 K. When all of the scattering processes were accounted for, the model showed the reduction of the thermal conductivity from 1.97 to 1.31 W/(cm · K) as the dislocation line density was raised from 10^{10} to 10^{12} cm⁻².

The same research team [10] also investigated separately the influence that is exerted by the doping density on the thermal conductivity. After making several modifications to the model, including the addition of a mixed dislocations component, and making corrections to some material parameters, the intrinsic thermal conductivity values were estimated at 3.36 and 5.40 W/(cm · K). When all of the scattering mechanisms were taken into account, the thermal conductivity fell off linearly with the natural logarithm of the doping density – about a factor of two decrease in κ for every decade increase in the doping density. As the latter was increased from 10^{17} to 10^{18} cm⁻³, the thermal conductivity dropped from 1.77 to 0.86 W/(cm · K).

Liu and Balandin [21] accounted for the three-phonon umklapp scattering, the scattering on impurity atoms and the scattering on external boundaries. The thermal conductivity was equal to 1.57 W/(cm · K) at room temperature.

Natural gallium (³¹Ga) consists of a mixture of two stable isotopes: 60.11 % ⁶⁹Ga and 39.89 % ⁷¹Ga. Natural nitrogen (⁷N) has also two stable isotopes: 99.63 % ¹⁴N and 0.37 % ¹⁵N. The isotope effect on the thermal conductivity of high-purity GaN was analyzed by Morelli *et al.* [14] with Callaway's model modified to include the contribution of both transverse and longitudinal phonons explicitly. For determining the combined scattering rate, the umklapp and normal processes, the phonon-isotope scattering and the scattering on boundaries were considered. The model predicted an isotope effect of 1.05 at 300 K.

This rather small value of the isotope effect was disputed by Lindsay *et al.* [22], who applied an *ab initio* (or “from first-principles”) formalism to determine the thermal conductivity of GaN defined by the three-phonon umklapp scattering and the phonon-isotope scattering. The first-principles method combines an exact iterative solution of the phonon Boltzmann transport equation in the single-mode relaxation time approximation with accurate computations of the second-order (harmonic) and third-order (anharmonic) interatomic force constants using the density functional theory and the density functional perturbation theory. The second-order interatomic force constants are required for the calculation of the phonon dispersion relation, while the third-order constants are necessary for the calculation of the three-phonon scattering rates. The advantage of this method is that no adjustable parameters are invoked as the interatomic force constants are the only input parameters required for the exact solution [23]. The thermal conductivity obtained by solving the phonon Boltzmann transport equation can be expressed as

$$\kappa = \frac{1}{\kappa_B T^2 N V} \sum_{\zeta} (\hbar\omega)^2 \bar{n}_{\zeta} (\bar{n}_{\zeta} + 1) v_{\alpha\zeta} d_{\beta\zeta}, \quad (16)$$

where N is the number of discretizing \vec{q} points in the Brillouin zone, V is the volume of the primitive unit cell, \bar{n}_{ζ} is the phonon group equilibrium population, v_{ζ} is the phonon group velocity, d_{ζ} is the mean free displacement, α and β are the Cartesian directions, ζ represents the phonon mode (\vec{q}, j) .

Unlike previous estimates [14, 24], Lindsay *et al.* showed that the isotope effect in wurtzite GaN is very large – about 1.65 at room temperature. The upper limit for κ was found to be around 4 W/(cm·K) at 300 K. These features were ascribed to the substantial isotope mixture in gallium, to the large frequency gap between the acoustic and optical phonon branches and to the high frequency scale of the phonon dispersion in GaN. The latter two points slacken the phonon-phonon scattering and, in particular, the scattering between the acoustic and optical phonons. Since the phonon-isotope scattering is not affected by temperature, starting from 500 K with decreasing temperature, the phonon-phonon scattering becomes weaker, causing the thermal conductivity to rise and making the scattering on isotopes more pronounced. At room temperature, the in-plane κ_{in} (along the [100] direction) and the out-of-plane κ_{out} (along the c -axis, or the [001] direction) values of the intrinsic thermal conductivity (κ_{pure}) were 4.01 and 3.85 W/(cm·K), respectively. The corresponding values for GaN with the naturally occurring gallium isotope concentrations ($\kappa_{natural}$) were 2.42 and 2.39 W/(cm·K).

The natural structure of GaN is wurtzite – a crystal structure belonging to the hexagonal crystal system, which is the most anisotropic among the other six crystal systems. Several investigations showed that the thermal conductivity of wurtzite GaN is isotropic around room temperature [24, 25]. On the other part, in most analytical works, the average value of the three directions or the value of one arbitrary direction are often taken as the thermal conductivity, omitting the anisotropy of the material. By performing non-equilibrium molecular dynamics simulation, Ju *et al.* [26] recently demonstrated that κ of defect-free wurtzite GaN along the [001] direction, is by a factor of 1.25 higher than that along [100] at room temperature. The evident anisotropic behavior of the thermal conductivity of wurtzite GaN at low temperatures was confirmed by Wu *et al.* [27], Qin *et al.* [28] and Jiang *et al.* [29]. In their research, Wu *et al.* developed a full-dispersion Monte Carlo simulation method coupled with the first-principles technique. According to their combined approach, the phonon dispersion relation and the scattering rates were computed using the first-principles method and served as the input data for the Monte Carlo solution of the phonon Boltzmann transport equation by applying the open-source ShengBTE code [30]. The thermal conductivity at room temperature of isotopically pure GaN along the [001] and [100] directions was calculated to be 4.04 and 3.62 W/(cm·K), respectively, leading to an anisotropy factor of 1.12. As the temperature decreased from 500 to 100 K, the factor increased from 1.10 to 1.25, indicating that the anisotropy of wurtzite GaN cannot be neglected, especially at low temperatures.

Qin *et al.* and Jiang *et al.* employed the first-principles method with a plane-wave pseudopotential formulation in the framework of the density functional theory as implemented in the Vienna Ab initio Simulation Package. The generalized gradient approximation with the Perdew–Burke–Ernzerhof parameterization was adopted for the exchange-correlation functional. The numerical solutions of the linearized phonon Boltzmann transport equation were also carried out using the ShengBTE software package. As was found by Qin *et al.*, the thermal conductivity at room temperature along the [001] and [100] directions of bulk GaN was evaluated to be 3.25 and 2.80 W/(cm·K), respectively, yielding an anisotropy factor of 1.16. In excellent agreement with the results obtained by Wu *et al.*, Jiang *et al.*, it determined the κ_{out} and κ_{in} values to be 4.09 and 3.63 W/(cm·K) at 300 K, respectively, giving an anisotropy factor of 1.13. When the temperature fell off from 800 to 100 K, the factor grew from 1.09 to 1.42.

By employing the first-principles approach to analyze the thermal conductivity of isotopically pure GaN, Garg *et al.* [31] recently observed a remarkable feature of the thermal conduction, namely, a spectral focusing of κ . It turns out that almost 60 % of the heat is transferred by phonons in a very narrow frequency range of 5–7 THz, which spans 9 % of the frequency range in GaN. The κ_{out} and κ_{in} values for the isotopically pure material were determined to be 3.73 and 3.90 W/(cm·K), respectively. The corresponding values for $\kappa_{natural}$ were equal to 2.34 and 2.38 W/(cm·K) – a very good agreement with the results of Lindsay *et al.*

Over the last few years, the application of machine learning techniques to solid-state physics have rapidly gained in popularity. The assessment of the thermal conductivity of bulk GaN using the innovative high-dimensional neural-network method developed by Behler and Parrinello [32] was first demonstrated by Minamitani *et al.* [33]. The thermal conductivity at room temperature along the [001] and [100] directions was computed to be 3.10 and 2.76 W/(cm·K), respectively. To estimate the accuracy of the method, these values were compared to the density functional theory calculation results, which were $\kappa_{\text{out}} = 3.26$ W/(cm·K) and $\kappa_{\text{in}} = 2.74$ W/(cm·K).

The dependence of the thermal conductivity on temperature for wurtzite GaN with naturally occurred isotope proportions obtained mainly by calculations is shown in Figure 1. For the sake of comparison, the experimental results of Jezowski *et al.* [25] and Simon *et al.* [34] are also included. The dislocation line density in “Kotchetkov *et al.* (2001)” is 10^{10} cm⁻². The dislocations in “Zou *et al.* (2002)” are evenly distributed among the screw, edge and mixed components. For “Jezowski *et al.* (2003)”, the best of the highly conductive samples is taken. The “Simon *et al.* (2014)” data points are given for the semi-insulating oxygen-magnesium co-doped sample. The “Minamitani *et al.* (2019) [001]” and “Minamitani *et al.* (2019) [100]” curves correspond to the κ values derived by the neural-network technique.

The dependence of the thermal conductivity on temperature for isotopically pure wurtzite GaN is shown in Figure 2.

Experimental Studies of the Thermal Conduction in GaN. The earliest experimental study of the thermal conduction in GaN was carried out by Sichel and Pankove [35] for a temperature range from 25 to 360 K. The bulk sample was fabricated by HVPE on a sapphire substrate [36]. The process included a two-hour reaction of gallium monochloride with ammonia at 1223 K. After the substrate was removed, the two surfaces of the GaN layer with a thickness of 400 μm were polished flat and parallel. The thermal conductivity evaluated along the *c*-axis had a slope of -0.76 around room temperature and a value of 1.3 W/(cm·K). Both of these values were small in comparison to the contemporary estimates [13], which was attributed by the authors to the high impurity density.

Using a scanning thermal microscope, Asnin *et al.* [37] performed high spatial resolution measurements of the thermal conductivity at 300 K on different patterned sections of GaN prepared by the lateral epitaxial overgrowth (LEO) technology on a sapphire substrate. The samples were fabricated by metalorganic chemical vapor deposition (MOCVD) at 1273 K with trimethylgallium and ammonia as the source precursors. On several sections of GaN, values of 1.7–1.8 W/(cm·K) were registered. Since the quality of the material obtained by MOCVD is generally better than that of the corresponding HVPE

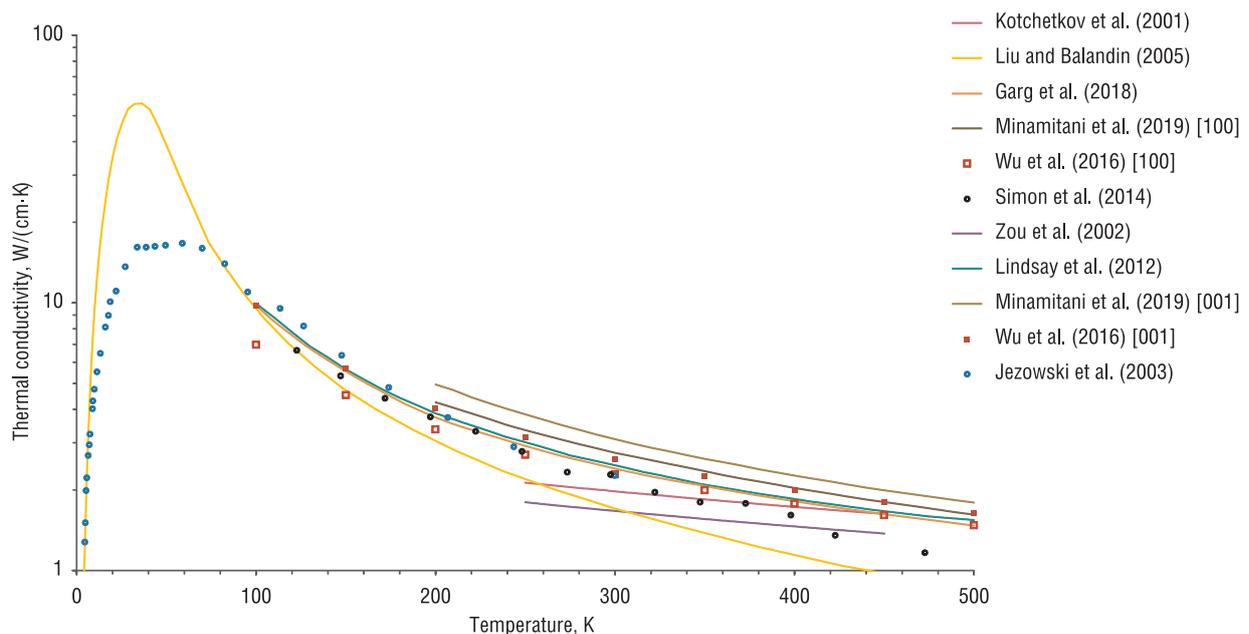


Figure 1. Thermal conductivity of wurtzite GaN with naturally occurred isotope proportions as a function of temperature obtained mainly by calculations

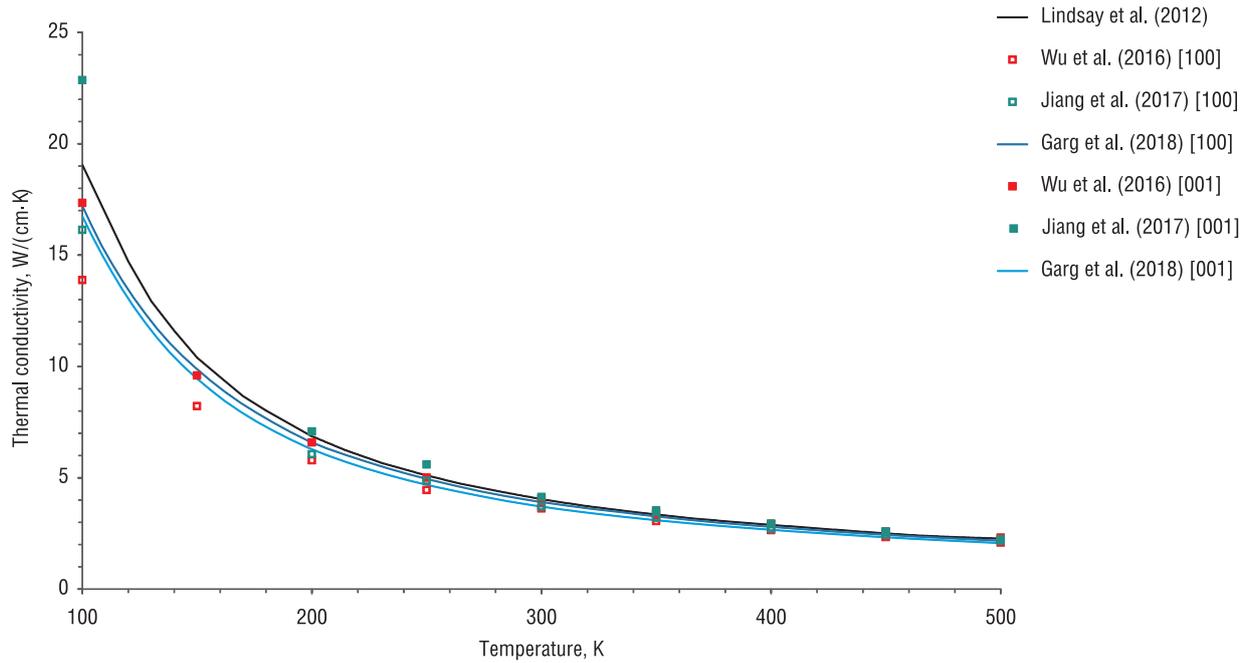


Figure 2. Thermal conductivity of isotopically pure wurtzite GaN as a function of temperature

sample, the higher values of κ were believed to be an indication of the reduced defect densities. Using the third-harmonic ($3-\omega$) electrical method, this assumption was soon corroborated by Luo *et al.* [38], who obtained a room temperature thermal conductivity in excess of 1.55 W/(cm·K) for the LEO films grown by MOCVD at 1333 K. In the paper [39], a comparison was drawn between the fully and the partly coalesced LEO sections of GaN prepared by MOCVD at 1273 K on a sapphire substrate. The measurements were taken by a scanning thermal microscope having a spatial resolution of $\sim 2-3 \mu\text{m}$. On the fully coalesced sample, the thermal conductivity lay in an interval from 1.86 to 2.05 W/(cm·K) over a distance of approximately 50 μm . One of the partly coalesced samples had κ between 2.00 and 2.10 W/(cm·K).

The thermal conductivity of GaN as a function of the doping density was experimentally researched by Florescu *et al.* [15]. The three sets of samples were fabricated by HVPE in a vertical reactor. The process consisted of two stages. First, gallium monochloride was synthesized upstream by a reaction of hydrogen chloride gas with liquid gallium at 1073–1173 K. Second, the gallium monochloride was transported to a sapphire substrate where it reacted with ammonia at 1273–1373 K forming a GaN layer. In all three sets, the thermal conductivity reduced linearly with the natural logarithm of the doping density, the variation being about a factor of two decrease in κ for every 10-fold increase in the doping density. For one of the most lightly-doped sample ($6.9 \cdot 10^{16} \text{ cm}^{-3}$), the thermal conductivity was found to be 1.95 W/(cm·K) at 300 K. As it was mentioned above, the dependence of this kind would later be confirmed analytically by Zou *et al.* [10] in a framework of Callaway's model.

One of the common impurities that affect the thermal conductivity of GaN is oxygen, which enters the lattice not by the replacement of nitrogen atoms but by the incorporation of gallium vacancies. The metal vacancies are very effective phonon scatterers [13]. To verify this view, Slack *et al.* [24] considered the thermal conductivity of a GaN plate manufactured by HVPE on a sapphire substrate, which was subsequently deleted. The major impurities were oxygen ($2.1 \cdot 10^{16} \text{ cm}^{-3}$) and silicon ($3.7 \cdot 10^{15} \text{ cm}^{-3}$). The thermal conductivity was equal to 2.27 W/(cm·K) at 300 K and possessed a slope of -1.22 in a temperature range from 80 to 300 K.

The first systematic study of the low-temperature behavior of GaN was reportedly carried out by Jezowski *et al.* [25] by comparing highly conductive and highly resistive (doped by magnesium) single crystals. The free-standing bulk samples were fabricated from nitrogen solution in liquid gallium at a high temperature and under a high pressure. The measurements were performed by the axial stationary heat flow method in a temperature range 4.2–300 K. The best of the highly conductive samples showed a thermal conductivity of 2.26 W/(cm·K) at 300 K, while the magnesium-doped GaN layer had $\kappa = 1.60 \text{ W/(cm·K)}$.

The high-temperature thermal properties of GaN were studied by Shibata *et al.* [40]. The single crystal sample was fabricated in a conventional vertical HVPE reactor at 1223–1293 K on a sapphire substrate. The free-standing sample unintentionally doped with silicon ($2.1 \cdot 10^{17} \text{ cm}^{-3}$) was spontaneously separated from the substrate. The thermal conductivity along the *c*-axis was obtained for a temperature range from 300 to 850 K and was equal to $2.53 \text{ W}/(\text{cm} \cdot \text{K})$ at room temperature. This value was highest among the previously reported ones for bulk-shaped samples [24, 25, 35]. Another work on the high-temperature behavior of GaN was presented by Slomski *et al.* [11]. The two bulk samples were grown by HVPE on sapphire and by the ammonothermal method on GaN, with the substrates subsequently removed. The density of dislocations laid in a range from 10^4 to 10^6 cm^{-2} . Using the $3\text{-}\omega$ technique, the thermal conductivity was extracted for a temperature range 300–460 K. The room temperature values of κ were equal to 2.65 and $2.79 \text{ W}/(\text{cm} \cdot \text{K})$, respectively, and the slopes were determined to be -1.43 and -1.38 .

Further investigation of the influence that is exerted by impurities on the thermal conductivity of GaN was made by Jagannadham *et al.* [41], who examined undoped, *n*-type (silicon-doped) and *p*-type (magnesium-doped) films on a sapphire substrate. The samples were fabricated by MOCVD at 1303 K for different periods of time with trimethylgallium and ammonia as the precursors. The deposition process was common to all three films except the dopant incorporation. The $3\text{-}\omega$ measurements were taken at 215, 250 and 300 K. The values of κ – 5.4, 4.8 and $2.8 \text{ W}/(\text{cm} \cdot \text{K})$ – in the undoped film were much higher compared to those in the *n*- and *p*-type samples. This was due to the absence of the phonon scattering on dopants, the larger thickness of the undoped film and the correspondingly lower density of dislocations. The presence of silicon and hydrogen in the *n*-type films was responsible for the values lower than those in the *p*-type films doped only by magnesium. Simon *et al.* [34] investigated the impact of oxygen and oxygen-magnesium co-doping on the thermal conductivity of bulk crystals with a low dislocation density ($\sim 10^4 \text{ cm}^{-2}$) grown by the ammonothermal method. The $3\text{-}\omega$ measurements were performed between 120 and 520 K. The heavily oxygen-doped sample attained the lowest value of κ – $1.64 \text{ W}/(\text{cm} \cdot \text{K})$ at 300 K – which was ascribed to the increased phonon-dopant scattering. On the contrary, the highest value – $2.30 \text{ W}/(\text{cm} \cdot \text{K})$ at 300 K – was observed for the semi-insulating oxygen-magnesium co-doped sample.

A high thermal conductivity of $2.94 \text{ W}/(\text{cm} \cdot \text{K})$ was determined by Richter *et al.* [42] for a cylindrical sample of 3.6 mm thickness and 6.0 mm diameter manufactured by HVPE.

The thermal conductivity of wurtzite GaN as a function of temperature obtained primarily by measurements is presented in Figure 3. For the sake of comparison, the theoretical results of Liu and Balandin and Garg *et al.* are also given. The “Slomski *et al.* (2018)” curve has the slope of -1.38 .

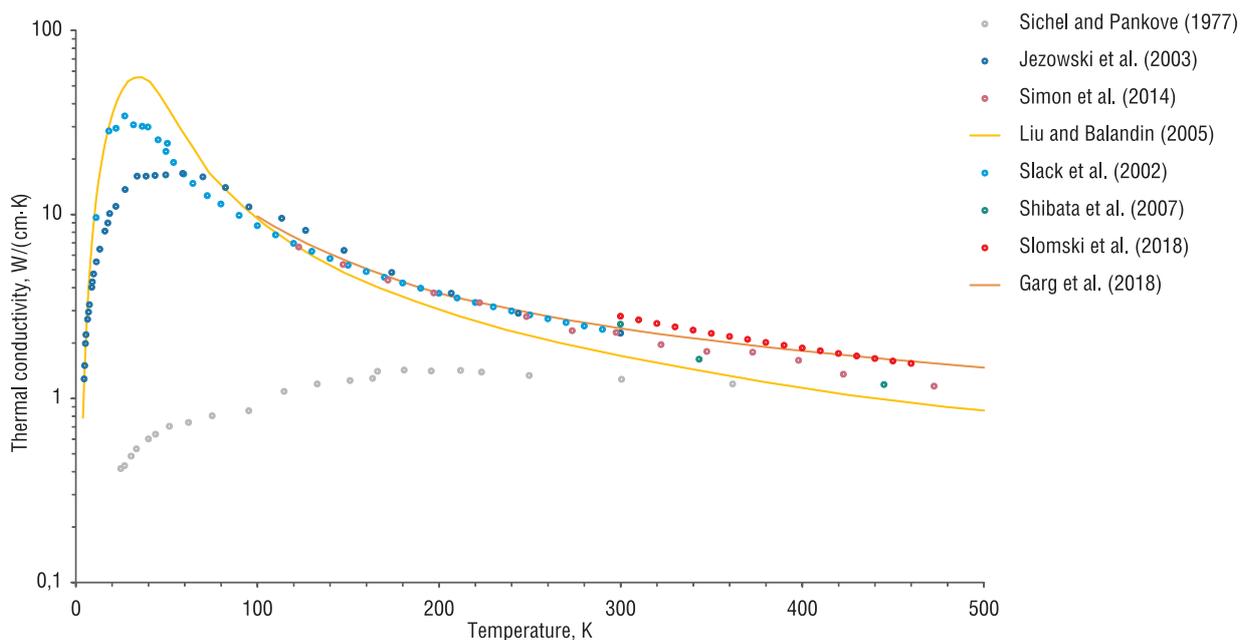


Figure 3. Thermal conductivity of wurtzite GaN as a function of temperature obtained primarily by experiment

Conclusions. In this review, a number of theoretical and experimental studies of the thermal conductivity of wurtzite GaN, particularly its dependence on temperature, has been described. A comparison of the results has also been made.

The heat in GaN is transported almost exclusively by phonons. Its thermal conductivity has a temperature behavior typical of most nonmetallic crystals: κ increases as T^3 at lower temperatures, comes up to a maximum at approximately $0.05\theta_D$ and decreases as T^{-1} at higher temperatures. At temperatures below θ_D , the thermal conductivity is limited by the scattering of the phonons on the external boundaries. As the temperature rises, the three-phonon umklapp scattering becomes predominant. The phonons in GaN can also be scattered by different point defects, such as impurity atoms, vacancies and isotopes.

For isotopically pure undoped wurtzite GaN, the value of κ at room temperature has been estimated as high as 5.4 W/(cm·K). The thermal conductivity of the films prepared by the LEO technology using MOCVD has been determined experimentally to be in a range from 1.55 to 2.10 W/(cm·K) at 300 K. The maximum room temperature value measured for the bulk-shaped samples of single crystal GaN has been 2.79 W/(cm·K).

The anisotropy of the thermal conductivity of GaN has remained to be a subject of discussion. While some investigators have shown that κ is isotropic at around room temperature, others have affirmed that this parameter exhibits a noticeable anisotropy, especially at low temperatures.

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Information about the authors

Vladislav S. Volchek – Researcher, Belarusian State University of Informatics and Radioelectronics (6, P. Brovka Str., 220013, Minsk, Republic of Belarus). <https://orcid.org/0000-0001-6942-810X>. E-mail: vlad.volchek@bsuir.by

Maryia S. Baranova – Researcher, Belarusian State University of Informatics and Radioelectronics (6, P. Brovka Str., 220013, Minsk, Republic of Belarus). <https://orcid.org/0000-0002-2618-4464>. E-mail: baranova@bsuir.by

Viktor R. Stempitsky – Ph. D. (Engineering), Associate Professor, Vice-Rector for Research and Development, Head of Research and Development Department, Belarusian State University of Informatics and Radioelectronics (6, P. Brovka Str., 220013, Minsk, Republic of Belarus). <https://orcid.org/0000-0001-9362-7539>. E-mail: vstem@bsuir.by

Информация об авторах

Волчэк Владислав Сергеевич – научный сотрудник, Белорусский государственный университет информатики и радиоэлектроники (ул. П. Бровки, 6, 220013, Минск, Республика Беларусь). <https://orcid.org/0000-0001-6942-810X>. E-mail: vlad.volchek@bsuir.by

Баранова Мария Сергеевна – научный сотрудник, Белорусский государственный университет информатики и радиоэлектроники (ул. П. Бровки, 6, 220013, Минск, Республика Беларусь). <https://orcid.org/0000-0002-2618-4464>. E-mail: baranova@bsuir.by

Стемпіцкіі Віктор Романавіч – кандидат технических наук, доцент, проректор по научной работе, начальник научно-исследовательской части, Белорусский государственный университет информатики и радиоэлектроники (ул. П. Бровки, 6, 220013, Минск, Республика Беларусь). <https://orcid.org/0000-0001-9362-7539>. E-mail: vstem@bsuir.by