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On the upper limit of thermal conductivity GaN crystals

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Abstract

The maximal value of thermal conductivity κ_{max} of the perfect wurtzite GaN crystal containing isotopes of natural abundance is estimated. Our upper limit of $\kappa = 4800$ W/K m at $T_{max} = 32$ K is smaller than that calculated by Liu and Balandin $\kappa = 6000$ W/K m and higher than that obtained by Slack et al. $\kappa = 3750$ W/K m. The phenomenological dependence $\kappa \propto T^{-1.43}$ obtained by Mion et al. for the temperature interval 300–450 K is extended to 200–300 K. For temperatures higher than T_{max} the best fitting of our experimental data to Callaway's formula is obtained for Grueneisen's constant equal to $\gamma = 1.35$.

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In heterostructures of GaN/AlGaN in electric fields of the order of 100 kV/cm, the density of dissipated power reaches 10^6 W/cm^2 [1–3]. This means that gallium-nitride-based devices suffer from heating effects that significantly limit the performance of high electron mobility transistors and laser diodes thereby reducing the device's life and reliability. This means that the accurate knowledge of the gallium nitride thermal conductivity is crucial to understanding thermal effects. For this reason, the investigation of the temperature behavior of thermal conductivity coefficient is an actual and reasonable problem.

Recently four groups measured thermal conductivity of GaN in the wide temperature ranges. Luo et al. [4] have measured the thermal conductivity of both LEO and HVPE single crystals of GaN over the temperature range 60–300 K. Slack et al. [5,6] performed measurements in the temperature interval 11–300 K. We measured thermal conductivity of massive high pressure growth dislocation-free crystalline sample containing about 10^{20} per cm^{-3} point defects in the temperature interval 4.5–300 K and performed a preliminary analysis of temperature dependence of it [7]. We also measured heat capacity of GaN for temperatures between 5 and 300 K, and as a result we established the value of Debye's temperature of GaN [8]. These measurements also allow us to obtain the temperature dependence of the phonon mean free path L_{eff}. Measurements of thermal conductivity for temperatures higher than 300 K were performed by Mion et al. [9].

Slack et al. compared their experimental results to the Callaway formula considering separately the contributions of longitudinal and transverse phonons. Therefore, their fitting procedure relays on six fitting parameters, namely two Debye's temperatures θ_L, θ_T Grueneisen's constants γ_L, γ_T and constants b_L, b_T related to the Umklapp processes (L and T stands for longitudinal and transverse phonons, respectively). The single set of their free adjustable parameters contained also the relaxation rate for the normal processes τ_N^{-1}. Liu and Balandin [10] considered thermal conductivity GaN. They argued that in the case of GaN specimens the normal processes do not influence the thermal conductivity because of strong scattering on point defects. For this reason we use Callaway's formula with $\kappa_2 = 0$. The results of measurements of thermal conductivity on wurtzite GaN samples [4–7] were analyzed by Kamatagi et al. [11] employing Holland's and

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modified Callaway's models. A good representation of the temperature dependence of κ was obtained with a single set of phonon–phonon scattering parameters. The purpose of this study is different from Kamatagi et al. [11]. Having in mind the needs of high-power electronic devices manufactured from GaN we shall establish the *upper* limit of thermal conductivity for $T > T_{\max}$. We base our approach on the standard Callaway formula and use only *two* free adjustable parameters.

In dielectric and semiconducting crystals heat is carried mainly by acoustic phonons. As a rule, the group velocities of such phonons are much greater than those of optical phonons. Therefore, heat flow in semiconducting crystals is limited by various mechanisms of scattering of acoustic phonons. There are two principal kinds of phonon scattering in crystals, namely resistive and normal processes [12]. They are characterized by the corresponding relaxation times τ_{R} and τ_{N}. Normal processes do not change the total momentum of phonons. Such processes influence the heat flow only indirectly. The resulting phonons of higher energy are scattered more intensively on point defects and have greater probability of participating in Umklapp processes. In the perfect crystals (dislocation and chemical impurity-free samples), the relevant intrinsic resistive scattering is due to three-phonon Umklapp with relaxation time τ_{U}, the point defect scattering with characteristic time τ_{P}, and the boundary scattering τ_{B}. The characteristic time τ_{P} is determined mostly by the crystal's natural isotope composition.

The total thermal conductivity $\kappa(T)$ can be written as [13]

$$\kappa = \kappa_1 + \kappa_2, \tag{1}$$

where $\kappa_1(T)$ and $\kappa_2(T)$ are defined by

$$\kappa_1(T) = \left(\frac{k_{\mathrm{B}}}{\hbar}\right)^3 \frac{k_{\mathrm{B}}}{2\pi^2\bar{v}} T^3 \int_0^{\frac{\theta_D}{T}} \frac{\tau_c x^4 \mathrm{e}^x}{(\mathrm{e}^x-1)^2}\mathrm{d}x, \tag{2}$$

$$\kappa_2(T) = \left(\frac{k_{\mathrm{B}}}{\hbar}\right)^3 \frac{k_{\mathrm{B}}}{2\pi^2\bar{v}} T^3 \left\{\int_0^{\frac{\theta_D}{T}} \frac{\tau_C}{\tau_N} x^4 \mathrm{e}^x \left(\mathrm{e}^x-1\right)^{-2}\mathrm{d}x\right\}^2 \times \left[\int_0^{\frac{\theta_D}{T}} \frac{\tau_C}{\tau_N\tau_R} x^4 \mathrm{e}^x \left(\mathrm{e}^x-1\right)^{-2}\mathrm{d}x\right]^{-1}, \tag{3}$$

where k_{B} and $\hbar$ are Boltzmann's and Planck's constants respectively, $\bar{v}$ is the mean velocity of phonons, θ_D is Debye's temperature, T is the ambient temperature, $x = \hbar\omega/k_{\mathrm{B}}T$, ω is the phonon frequency, $\tau_C^{-1} = \tau_R^{-1} + \tau_N^{-1}$, and $\tau_R^{-1} = \tau_U^{-1} + \tau_P^{-1} + \tau_B^{-1}$.

When $\tau_N >> \tau_R$, the combined relaxation time $\tau_c \approx \tau_R$ and the term κ_2 (3) is negligibly small. The generally accepted expression for Umklapp phonon scattering rate is (cf. e.g. [5])

$$\tau_U^{-1} = \frac{\hbar\gamma^2 T\omega^2}{M\bar{v}^2\theta_D} \exp\left(-\frac{\theta_D}{bT}\right), \tag{4}$$

where γ is Grueneisen's constant, M is the mass of GaN molecule, and is b is a fitting parameter. As a rule for crystalline solids, $2 \le b \le 3$ [12].

For phonons scattered by isolated defects of mass different from that of the host in an otherwise perfect crystal, Tamura derived a scattering rate [14]

$$[\tau_P(x)]^{-1} = \frac{V k_{\mathrm{B}}^4 \tilde{g}}{12\pi\,\hbar^4\,\bar{v}^3} x^4 T^4, \tag{5}$$

where V is the volume of the elementary cell, the parameter $\tilde{g}$ is

$$\tilde{g} = \frac{\sum_\sigma g(\sigma)\bar{M}^2(\sigma)}{\left[\sum_{\sigma'} \bar{M}(\sigma')\right]^2}, \tag{6}$$

with $g(\sigma)$ is the constant which express the strength of isotope effects. It depends on mass $M_i(\sigma)$ and fraction $f_i(\sigma)$ in the ith isotope of σ atom as

$$g(\sigma) = \sum_i f_i(\sigma)\left[1 - \frac{M_i(\sigma)}{\bar{M}(\sigma)}\right]^2. \tag{7}$$

Above $\bar{M}(\sigma) = \sum_i f_i(\sigma) M_i(\sigma)$ is the average mass of σ atom. The boundary scattering is important for low temperatures and does not influence thermal conductivity in the intermediate and high temperatures. The related scattering rate τ_B^{-1} is

$$\tau_B^{-1} = \frac{\bar{v}}{L_{\mathrm{eff}}}, \tag{8}$$

where L_{eff} is the effective phonon mean free path. For $T \ll \theta_D$ it is of the order of the cross-sectional dimensions. A precise determination of L_{eff} is crucial for correctly describing the $\kappa(T)$ curve in the low temperature range.

As we have recently established, for GaN, the Debye temperature $\theta = 365$ K [8], which is lower than used by other authors [5,6,10,15,16]. It is therefore obvious that one should revise the fitting procedure. We calculate $\tilde{g}$ (Eq. (6)) accounting for natural composition of isotopes of Ga and N. Our calculations show, in agreement with Ref. [6], that the contribution of nitrogen isotopes to $\tilde{g}$ is ten times smaller than the contribution of isotopes of gallium.

Since other defects always present in available specimens diminish the thermal conductivity coefficient, we obtain the upper limit of thermal conductivity of GaN specimens with the natural composition of isotopes. The Grueneisen parameter for hexagonal GaN was calculated by Łopuszyński and Majewski [17]. Their value $\gamma = 1.28$ is bigger that used by other authors [6,10,18]. For temperatures higher than $T_{\max}$ (corresponding to $\kappa_{\max}$), the best fitting is obtained for $\gamma = 1.35$, which is close to the calculated value [17]. As a result of fitting the Callaway formula to our experimental results (with $\tau_N \gg \tau_R$), we found $b = 2.5$. In Table 1 we collected the values of the above parameters.

Table 1
Single parameter set based on Callaway's model used to represent the temperature dependence of the thermal conductivity κ

$\bar{v}$ (cm/s)	γ	$\tilde{g}$	b	θ_D (K)
5.1×10^5	1.35	1.37×10^{-4}	2.5	365