



BUILT-IN-POLARIZATION FIELD EFFECT ON INTRINSIC AND EXTRINSIC THERMAL CONDUCTIVITY OF GAN AND INN

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ABSTRACT

We have investigated theoretically the effect of built-in-polarization field on thermal conductivity of bulk Wurtzite GaN and InN. Built-in-polarization field modifies thermal parameters and combined scattering relaxation time. Using these modified parameters, we have calculated intrinsic and extrinsic thermal conductivities of GaN and InN for with and without polarization. We have also observed that, increment in intrinsic thermal conductivities of GaN and InN is 17.75 %, and 23 % respectively, and in extrinsic thermal conductivities of GaN and InN is 11.87 % and 11.79 % respectively, because of built-in-polarization field. The theoretical analysis shows that up to a certain temperature the polarization effect acts as negative effect, and reduces the intrinsic and extrinsic thermal conductivities. However after this transition temperature both thermal conductivities are significantly enhanced. The method we have developed can be taken in to account during the simulation of heat transport in optoelectronic nitride devices to minimize the self-heating processes.

Keywords: Built-In-Polarization, Phonon Scattering, Relaxation Time, Self-Heating and Thermal Conductivity

I. INTRODUCTION

Group III Wurtzite nitride semiconductor are promising compounds for light emission in the green to the near-ultraviolet (UV) spectral range. These compounds have potential to extend this range further into the UV region and widely use in LASER, LED, solar cell, and other optoelectronic devices. One of the important reasons for broad applications of AlN, GaN and InN is direct wide bandgap, which covers ultraviolet to infrared energy range ($E_{g,AlN} = 6.2$ eV, $E_{g,GaN} = 3.4$ eV, $E_{g,InN} = 0.64$ eV).[1] The performance of Wurtzite nitride devices is reliable, but affected by the self-heating.[2] The cause of self-heating is poor heat removal at the active region of the device. The heat removal from active region directly depends on thermal transport property of the material. Thus, properties of material that can enhance thermal transport should be explored. Intentionally introducing this property in the material, the high thermal conductivity as desired can be achieved. The high thermal conductivity can minimize self-heating effect and can improve the performance as well as lifetime of the nitride devices [3].

A number of groups have studied the thermal conductivity of binary nitrides theoretically and experimentally. Gallium Nitride (GaN) is the key material for the next generation of high frequency and high power transistors, which can be operated at high temperature. A. Witek have predicted the intrinsic thermal

conductivity of GaN, is $410 \text{ Wm}^{-1}\text{K}^{-1}$ [4]. The thermal conductivity for isotopic alloy enriched GaN is found to be $400 \text{ Wm}^{-1} \text{ K}^{-1}$ [3], which is theoretically predicted. Kotchetkov et al. have extensively calculated the effect of dislocation and isotope scattering in GaN [5]. Asnin et al. measured the room temperature thermal conductivity of GaN film and reported $180 \text{ Wm}^{-1}\text{K}^{-1}$ [6]. Slack et. al. in 2002 [7] predicted the intrinsic thermal conductivity in absence of isotopic scattering at room temperature is $250 \text{ Wm}^{-1} \text{ K}^{-1}$. The thermal conductivity of highly conducting n-type GaN found as $227 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature [8]. Liu et. al. showed that the thermal conductivity in free standing GaN films to be as high as $225 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature [9].

The discovery of narrow band gap of InN in 2002 [10] open vigorous research field to study the thermal conductivity of InN. The first experimental measurement of thermal conductivity was $45 \text{ Wm}^{-1}\text{K}^{-1}$, reported by Krukowski et al [11], which was grown by nitrogen microwave plasma chemical vapor deposition including point defects and grain boundary scattering. This thermal conductivity is much smaller than the thermal conductivity $176 \text{ Wm}^{-1}\text{K}^{-1}$ which is estimated by phonon-phonon scattering. Sahoo et al [12] predicted the thermal conductivity of InN is $132 \text{ Wm}^{-1}\text{K}^{-1}$ by taking phonon-phonon, mass difference, dislocation and phonon-electron scattering rate at room temperature. Another experimental value of thermal conductivity k is $120 \text{ Wm}^{-1}\text{K}^{-1}$ reported by Levander et al [13], for high quality irradiated InN film.

The theoretical and experimental studies of thermal conductivity have taken care of different growth techniques, various phonon scattering mechanisms and defects of AlN material [4,5]. One of the most important and peculiar property of binary nitrides is strong built-in-polarization [14–20]. Right from the beginning, a majority of experiments and theoretical calculations have been devoted to explore polarization properties for the major WZIII–V nitrides. The experimental evidences are the red shift of transition energy [21], the high carrier density at hetero junctions [14], reduction of the oscillator strength in AlGaIn/GaNQW [22], etc. Leroux et al. [23] have deduced the built-in-electric field by conducting time dependent photoluminescence (PL) experiments on AlGaIn/GaN QWs. Hoggetal. [24] carried out cathode luminescence (CL) measurements on a series of AlGaIn/GaNQW under different strain conditions and art fully determined there lative effects of piezoelectric (PZ) and spontaneous (SP) polarization. Simon etal. [25] performed PL experiments at 2K for a series of GaN/AlGaIn QWs with different aluminum composition x and well thickness and showed that the measured electric fields are larger than what is expected based on piezoelectric effects alone, which becomes direct experiment alp roof of the existence of important spontaneous polarization effects. O. Ambacher etal. [14–16] have experimentally determined the effect of polarization on 2DE Gin hetero structures. The oretical studies have been done to account for the polarization effects in WZIII–V QWs. Bernardini etal. [17– 20] have calculated the SP and PZ constants of III–V nitrides by ab initio method. Liu etal. [26] have investigated the thermoelectric effects in wurtzitenit rides. Park and coworkers have investigated the oretically the SP and PZ effects in AlGaIn/GaNQWs [22].

To the best of our knowledge, thermal conductivity studies of binary nitrides have not included the built-in-polarization effect. The purpose of this study is to report the effect of built-in-polarization on intrinsic and extrinsic thermal conductivities of binary nitrides. The thermal conductivity of a “perfect” defect-free semiconductor is termed as intrinsic thermal conductivity. The extrinsic thermal conductivity is observed in the experiment where as intrinsic thermal conductivity gives maximum theoretical limit achievable for a “perfect” defect-free semiconductor. This sets the theoretical upper limit to thermal conductivity of binary nitrides. The

paper is organized in the following manner. The built-in-polarization, effective elastic constant and thermal conductivity are discussed in Section 2. The results and discussion is presented in Section 3. Conclusion is given in Section 4.

II. THE MODEL

2.1 Thermal conductivity

III-V nitrides crystalizes in wurtzite structure which is non-centro symmetric in nature. Due to lattice mismatch strain is produced. The non-centro symmetric nature and strain create built-in-polarization field (sum of spontaneous and piezoelectric polarization fields) [16]. In binary nitrides, the built-in-polarization field has only one non-vanishing component. This component points along the growth direction of the material i.e., along the z axis of the unit cell of the binary nitrides. For binary nitrides the built-in- polarization field along the z axis is given by [26]

$$p = p^{sp} + 2e_{13}\epsilon_1 + e_{33}\epsilon_3 \quad (1)$$

Here ϵ is strain and e_{kl} are piezoelectric constants. ϵ and e_{kl} are used to describe the piezoelectric properties of group III nitrides. In semiconductors, heat energy is carried by phonons. The phonons are influenced by built-in- polarization field. The compressive strain in the binary nitrides induces an increase in the elastic constants via nonlinear elasticity effects and tends to increase the sound velocity, the concomitant reduction of unit cell volume results in an increase of the density and hence lowers the sound velocity. The effective elastic constant is given by [27-30]

$$\bar{C}_{44} = C_{44} + [(e_{15}^2 + e_{31}^2 + e_{33}^2 + p_{sp}^2) / \epsilon_0 \epsilon] \quad (2)$$

The values of material parameters and polarization constants of binary nitrides are listed in Table1 [31-32]. The velocity of phonons is $v = (C_{44} / \rho)^{1/2}$. With the inclusion of built-in-polarization, the velocity becomes $v_{T,P} = (\bar{C}_{44} / \rho)^{1/2}$. The Debye frequency and Debye temperature are $\nu_D = v(3N/4\pi V)^{1/3}$ and $\theta_D = h \nu_D / k_B$. Here ρ , N and V are density, no. of atoms and volume of unit cell of binary nitrides, respectively.

Phonons are scattered due to defects, impurities and dislocation present in the binary nitrides. The combined phonon relaxation time s ($1/\tau_c$) are functions of phonon velocity, Debye frequency and Debye temperature. The important phonon scattering process are: normal scattering (τ_n), Umklapp scattering (τ_u), mass-difference scattering (τ_m), dislocation scattering (τ_d) and phonon-electron scattering (τ_{ph-e}). The combined phonon relaxation time ($1/\tau_c$) can be obtained by the summation of these scattering processes and is given by [31-33]

$$\frac{1}{\tau_c} = \frac{1}{\tau_n} + \frac{1}{\tau_u} + \frac{1}{\tau_m} + \frac{1}{\tau_d} + \frac{1}{\tau_{ph-e}} \quad (3)$$

The built-in-polarization field affects the elastic constant, phonon group velocity, Debye frequency, Debye temperature, phonon scattering relaxation time and combined scattering relaxation time of GaN and InN [31].

The total thermal conductivity is defined by electron and phonon component of thermal conductivity, using electron transport model and phonon transport model respectively. The Boltzmann transport equation (BTE)



with relaxation time approximation is used to calculate electron component of thermal conductivity by electrical transport model and the Callaway model with virtual crystal approximation is used to calculate phonon component of thermal conductivity phonon transport model. The total thermal conductivity is given by [33]

$$k(T) = k(T)_{Lattice} + k(T)_{Electron} \tag{4}$$

In semiconductors, the dominant contribution to the room temperature thermal conductivity comes from phonons. Florescu *et al.* [34] experimentally determined that the electronic contribution to thermal conductivity in nitrides is about 1.5×10^{-3} W/cm K which is three orders of magnitude smaller than the typical value of the lattice thermal conductivity. Thus, here we neglect the electronic contribution to thermal conductivity. According to Callaway model [33], the lattice thermal conductivity of binary nitrides is given by

$$k(T)_{Lattice} = k(T)_1 + k(T)_2 \tag{5}$$

$$k(T)_1 = \left(\frac{k_\beta}{\hbar}\right)^3 \frac{k_\beta}{2\pi^2 v} T^3 \int_0^{\theta_D/T} \frac{\tau_c x^4 e^x}{(e^x - 1)^2} dx \tag{6}$$

$$k(T)_2 = \left(\frac{k_\beta}{\hbar}\right)^3 \frac{k_\beta}{2\pi^2 v} T^3 \frac{\left[\int_0^{\theta_D/T} \frac{\tau_c}{\tau_n} \frac{x^4 e^x}{(e^x - 1)^2} dx \right]^2}{\int_0^{\theta_D/T} \frac{\tau_c}{\tau_r \tau_n} \frac{x^4 e^x}{(e^x - 1)^2} dx} \tag{7}$$

where $x = \hbar\omega/k_\beta T$. The important parameters influencing k are the phonon group velocity v , Debye temperature θ_D and the combined relaxation time τ_c [31-33]. The thermal conductivity when the combined relaxation time takes Umklapp and normal relaxation time into account, $\tau_c = \tau_u + \tau_n$ and neglects the others contribution of relaxation time, is termed as intrinsic thermal conductivity [33]; and when combine relaxation time includes all major phonon resistive and normal scattering processes, thermal conductivity is called as extrinsic thermal conductivity [31]. In order to know the effect of built-in-polarization on thermal conductivity, the revised values of v, θ_D, ω_D and τ_c are to be used.

Table 1. Material Parameters and polarization constants of GaN and InN [31-32].

Parameter	GaN	InN
a (Å)	3.189	3.540
c (Å)	5.185	5.700
ϵ	9.5	15.3
ρ (Kg m ⁻³)	6150	6890
C_{33} (GPa)	4.05	2.24
C_{44} (GPa)	1.05	0.48
e_{15} (C m ⁻²)	-0.30	-0.57
e_{31} (C m ⁻²)	-0.55	-0.56
e_{33} (C m ⁻²)	1.12	1.09
p_{sp} (C m ⁻²)	0.0842	0.042



Γ	1.3×10^{-4}	1.3×10^{-4}
N_D (m ⁻²)	1.5×10^{14}	1.5×10^{14}
n_e (m ⁻³)	2.6×10^{24}	3.3×10^{24}
ϵ_1 (eV)	10.1	7.1
m^*	$0.22 \times 9.1 \times 10^{-31}$	$0.14 \times 9.1 \times 10^{-31}$

Table 2. Elastic constant, phonon velocity, Debye frequency and Debye temperature of AlN GaN and InN [31-32].

Parameters	without Polarization	with Polarization	% increase
GaN			
C_{44} (GPa)	105	125	19
v (m s ⁻¹)	4931	5281	7
ν_D (sec ⁻¹)	7.85×10^{13}	8.40×10^{13}	7
θ_D (K)	600	642.5	7
InN			
C_{44} (GPa)	48	61.5	28.1
v (m s ⁻¹)	3233.8	3571.8	10.45
ν_D (sec ⁻¹)	8.63×10^{13}	9.53×10^{13}	10.45
θ_D (K)	660	729	10.45

III. RESULTS AND DISCUSSION

3.1 Revised Parameters

First we have calculated the revised parameters of GaN and InN taking into account the built-in-polarization effect. The effective elastic constant \bar{C}_{44} for GaN and InN, is estimated using the polarization constants and elastic constants. In this work, we have used theoretical values for spontaneous polarization and for the piezoelectric coefficients e_{15} , e_{31} and e_{33} [32]. For spontaneous polarization, only a single set of theoretical values, and no experimental values, are available. For the piezoelectric coefficients, only theoretical values are available for InN. In the case of GaN, there is a significant discrepancy between the experimental and theoretical values; for consistency, and given the large uncertainty likely to exist for both the experimental and theoretical values, we have used the theoretical values for e_{15} , e_{31} and e_{33} . The maximum values of polarization constants reported till date is used in the calculation [32]. The used parameters are listed in Table 1. For GaN and InN, estimation shows that \bar{C}_{44} is increased due to built-in-polarization. Then the average phonon group velocity v_p and v i.e, velocity with and without polarization are calculated [31-32]. The velocity (v_p) with polarization is higher than velocity (v) without polarization. The Debye frequency and Debye temperature with polarization are also estimated. Table 2 shows the values of elastic constant, phonon group velocity, Debye frequency and Debye temperature of GaN and InN for with and without polarization. The percentage increase of parameters due to

polarization is also estimated. The percentage change is defined by $[(v_p - v)/v] \times 100$, where v_p is velocity of phonons with polarization and v is velocity without polarization.

3. 2 Combined relaxation time (τ_c):

The combined phonon relaxation time of GaN and InN is calculated by Eq. (3) for with and without polarization including τ_n phonon relaxation time due to normal scattering and τ_r the combined relaxation time due to all resistive processes in binary nitrides. The phonon relaxation time (τ_n) due to normal scattering process depends on phonon-phonon interaction, volume of unit cell, phonon velocity and phonon frequency, which is calculated as function of phonon frequency for with and without polarization at room temperature. The drop-off in phonon-phonon interaction and volume of unit cell increases the phonon relaxation time τ_n and the gain in phonon velocity also increases the phonon relaxation time τ_n . The used parameters in calculation of τ_n are listed in Table 2.

The combined phonon relaxation time τ_r due to all resistive processes is computed, which includes phonon relaxation time due to Umklapp scattering process (τ_u), mass-difference scattering process (τ_m), dislocation scattering process (τ_d) and phonon-electron scattering process (τ_{ph-e}). The phonon relaxation time of GaN and InN due to umklapp scattering process (τ_u) is estimated for with and without polarization at room temperature and it depends on Debye frequency, shear modulus and volume of unit cell and phonon frequency. The applied parameters in calculation of τ_u as function of phonon frequency, are listed in Table 2 for with and without polarization. The phonon relaxation time of GaN and InN ascribable by mass-difference scattering process (τ_m) is forecasted for with and without polarization, where it depends on phonon frequency, phonon velocity and volume of unit cell. The utilized parameters in reckoning of the relaxation time due to mass-difference scattering process are inclination from Table 2 for with and without polarization. The phonon relaxation time (τ_d) of GaN and InN due to dislocation scattering process is calculated for with and without polarization, where the relevant parameters are phonon frequency, phonon velocity and volume of unit cell. The used parameters are listed in Table 2 for calculation of phonon relaxation time (τ_d) due to dislocation scattering process at room temperature. Similarly, the combined phonon relaxation time (τ_c) of GaN and InN is defined (3) for with and without polarization. The all relaxation times due to different scattering processes are discussed above and we have found the built-in-polarization increases the all phonon relaxation times. The contribution of all relaxation times are used in calculation of combined phonon relaxation time (τ_c) and which is increased due to built-in-polarization effect at room temperature. It has been observed that the combined phonon relaxation time τ_c decreases with increasing the phonon frequency in both with and without polarization. The increment in combined phonon relaxation time (τ_c) arises due to the built-in-polarization effect.

3.3 Thermal Conductivity (k)

The thermal conductivity k of GaN and InN is estimated by taking into account the above revised parameters. The factors affecting thermal conductivity k are phonon group velocity (V), Debye temperature (θ_D) and combined relaxation time (τ_c). The phonon velocity and Debye temperature are increased due to polarization effect. The combined relaxation times, as discussed above is significantly increased due to polarization effect. The high Debye temperature extends the upper limit of thermal conductivity integration.

The intrinsic and extrinsic thermal conductivity of GaN and InN for with and without polarization is plotted in Fig. 1 and Fig. 2, as a function of temperature, respectively. In both Figure, the red solid and empty circles curves respectively, represents the intrinsic thermal conductivity of GaN and InN for with and without polarization. Figures shows that, temperature range from 20 ⁰K to 125 ⁰K in GaN and 20 ⁰K to 120 ⁰K in InN, the intrinsic thermal conductivity without polarization is comparatively higher than intrinsic thermal conductivity with polarization, but after 125 ⁰K in GaN and 120 ⁰K in InN, the intrinsic thermal conductivity with polarization is vastly contributed by polarization. The maximum intrinsic thermal conductivity with polarization is 494 $Wm^{-1}K^{-1}$ at temperature 270 ⁰K in GaN and 514 $Wm^{-1}K^{-1}$ at temperature 280 ⁰K in InN. However, maximum intrinsic thermal conductivity without polarization is 418 $Wm^{-1}K^{-1}$ at temperature 250 ⁰K in GaN and 401 $Wm^{-1}K^{-1}$ at temperature 260 ⁰K in InN. These are the maximum thermal conductivity of GaN and InN in absence of defect, dislocation and phonon-electron scattering. At room temperature, the intrinsic thermal conductivities of GaN and InN for with polarization is 490 $Wm^{-1}K^{-1}$ and 513 $Wm^{-1}K^{-1}$ respectively, and for without polarization is 407 $Wm^{-1}K^{-1}$, 393 $Wm^{-1}K^{-1}$ respectively. This result shows that the increment in intrinsic thermal conductivities of GaN and InN is 17.75 % and 23 % respectively.

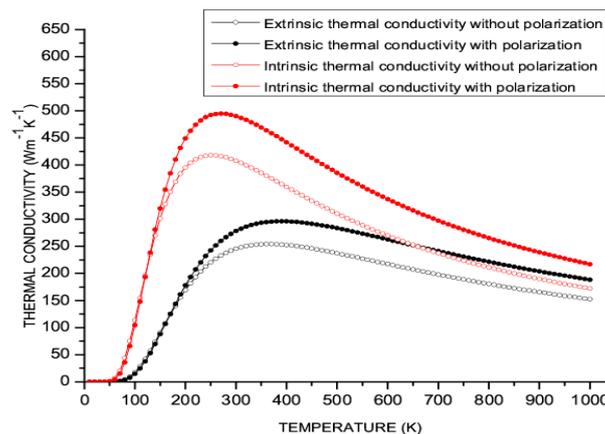


Fig. 1. Intrinsic and Extrinsic Thermal conductivity of GaN as a function of temperature for with and without polarization.

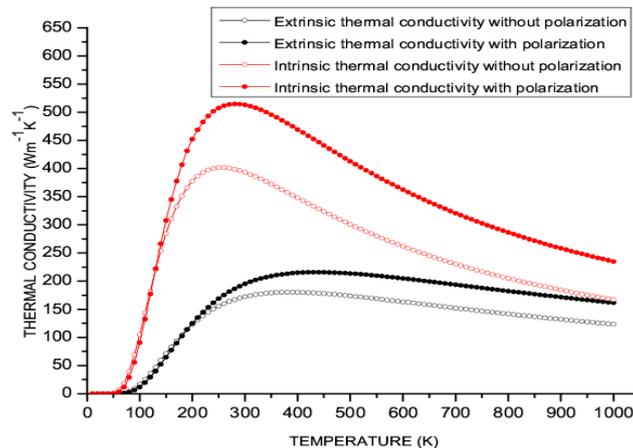


Fig. 2. Intrinsic and Extrinsic Thermal conductivity of InN as a function of temperature for with and without polarization.

Again, in Fig 1 and Fig 2, the black solid and empty circles curves respectively represent the extrinsic thermal conductivity of GaN and InN for with and without polarization, as a function of temperature. From the figures, it is clear that the extrinsic thermal conductivity of GaN and InN with polarization is less than without polarization from temperature range 20 °K to 165 °K in GaN and 20 °K to 190 °K in InN. The transition temperature is 165 °K in GaN and 195 °K in InN, for extrinsic thermal conductivity. Thus, polarization diminishes the extrinsic thermal conductivity up to temperature 165 °K in GaN and 195 °K in InN. After these temperature extrinsic thermal conductivity is vastly contributed by polarization and becomes maximum 296.23 $\text{Wm}^{-1} \text{K}^{-1}$ at temperature 390 °K in GaN and 215 $\text{Wm}^{-1} \text{K}^{-1}$ at temperature 440 °K in InN. However, the maximum thermal conductivity without polarization is 254.15 $\text{Wm}^{-1} \text{K}^{-1}$ at temperature 360 °K in GaN and 180 $\text{Wm}^{-1} \text{K}^{-1}$ at temperature 390 °K in InN. At room temperature, the extrinsic thermal conductivities of GaN and InN for without polarization is 245 $\text{Wm}^{-1} \text{K}^{-1}$ and 172 $\text{Wm}^{-1} \text{K}^{-1}$ respectively, and for with polarization is 278 $\text{Wm}^{-1} \text{K}^{-1}$ and 195 $\text{Wm}^{-1} \text{K}^{-1}$ respectively. Thus the increment in extrinsic thermal conductivities of GaN and InN is 11.87 % and 11.79 % respectively. This shows that polarization property enhances the room temperature thermal conductivity of GaN and InN.

It is reported in literature that built-in-polarization has negative effect on optical and electronic properties of the wurtzite nitrides [14-16]. Here we found that polarization has negative effect on extrinsic thermal conductivity up to a temperature 165 °K in GaN and 195 °K in InN and after these temperature extrinsic thermal conductivity is significantly contributed by built-in-polarization property [31]. This gives the idea of temperature dependence of built-in-polarization effect which signifies pyro-electric character of GaN and InN.

IV. CONCLUSION

We have studied theoretically the influence of built-in-polarization on intrinsic and extrinsic thermal conductivity of Wurtzite GaN and InN. We have found that built-in-polarization contributes to the effective elastic constant of binary nitrides and enhances the values of phonon group velocity, Debye frequency and Debye temperature. High phonon velocity lowers the phonon relaxation times and makes combined relaxation

time longer. High Debye temperature extends the upper limit of thermal conductivity integration. This extension gives a positive contribution to the thermal conductivity. In this work we have calculated the maximum thermal conductivity of GaN and InN taking only Normal and Umklapp scattering process, because at that condition the phonons scattered by phonons only and other impurity dislocation and electron scattering ignored in with and without polarization. The maximum thermal conductivity with polarization is $494 \text{ Wm}^{-1}\text{K}^{-1}$ at temperature $270 \text{ }^{\circ}\text{K}$ in GaN and $514 \text{ Wm}^{-1}\text{K}^{-1}$ at temperature $280 \text{ }^{\circ}\text{K}$ in InN. However, maximum thermal conductivity without polarization is $418 \text{ Wm}^{-1}\text{K}^{-1}$ at temperature $250 \text{ }^{\circ}\text{K}$ in GaN and $401 \text{ Wm}^{-1}\text{K}^{-1}$ at temperature $260 \text{ }^{\circ}\text{K}$ in InN. Our theoretical analysis shows that up to a certain temperature the polarization effect acts as negative effect and reduces the thermal conductivity but after this temperature the thermal conductivity is significantly contributed by the polarization effect. This shows the temperature dependence of the polarization effect and signifies the pyro-electric nature of GaN and InN. This method give us the idea to find the pyro-electric property of Nitride material. Thus, by intentionally introducing built-in-polarization mechanism in nitrides, high thermal conductivity value as desired can be achieved. The high thermal conductivity of GaN and InN has importance in its technological use as a heat-sink substrate for semiconductor devices. Future experiments should address built-in-polarization to confirm this predicted result. This is an important observation that should be taken into account in simulating heat transport in optoelectronic nitride devices to minimize the self-heating processes.

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