Polarization electric field effect on cross-plane thermal conductivity of GaN/In\textsubscript{x}Ga\textsubscript{1-x}N/GaN superlattice (x ≤ 0.3)

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Abstract. GaN/In\textsubscript{x}Ga\textsubscript{1-x}N/GaN superlattice (SL) has electric field at interfaces termed as interfacial polarization electric (IPE) field. In this work, effect of this electric field on cross-plane thermal conductivity (k\textsubscript{cp}) of the SL is investigated for Indium content x ≤ 0.3. IPE field revises phonon velocity which enhances interfacial scattering and thermal boundary resistance (TBR). This is due to unequal changes in specific heat and phonon velocity which leads to decrease in phonon transmission and more mismatches of acoustic properties of material. This reduces k\textsubscript{cp}. Room temperature k\textsubscript{cp} in presence (absence) of IPE field for GaN (10nm)/In\textsubscript{x}Ga\textsubscript{1-x}N (5nm) SL are 4.652 (5.720) and 4.282 (5.221) Wm\textsuperscript{-1}K\textsuperscript{-1} respectively, for x=0.1 and 0.3. This work demonstrates that electric field of nitride SL can be utilized to reduce k for optimum thermoelectric power production.

1. Introduction

Efficiency of a thermoelectric (TE) material is generally quantified by dimensionless figure of merit (ZT) which is directly proportional to electrical conductivity (σ) and Seebeck coefficient (S); but inversely proportional to k of the material [1]. The main paradigm to achieve material’s high ZT is to enhance their power factor (S\textsuperscript{2}σ) and/or reduce their k. A small reduction in k can improve TE efficiency significantly. Various approaches have been undertaken to reduce k of SL. Doping, alloying, period width variation, lattice mismatch, nano-particle substitution, interfacial roughness and atomic disorder etc [2-5]. Many of these studies have proved that k of SL is lower than the corresponding k of bulk material. Thus, more approaches must be explored to reduce k of SL. In this work, polarization property of nitride SL has been exploited to reduce k of SL.

Recent study reported that GaN /In\textsubscript{x}Ga\textsubscript{1-x}N/ GaN SL can be a promising contender for TE devices due to its lowest k within the III-V nitride alloys [6]. Its ZT can approach best TE material Bi and Te. This motivates to explore methods to enhance ZT of GaN /In\textsubscript{x}Ga\textsubscript{1-x}N SL reducing k and increasing σ , S of the SL further. This SL has a unique advantage called polarization mechanism which produces an interfacial polarization electric (IPE) field at each interface of the SL due to difference in spontaneous (SP) and lattice mismatch piezoelectric (PZ) polarizations of layers [7]. Szein et al. [6] demonstrated that IPE field of GaN/AlN/AlGaN SL improves σ and S resulting in high ZT. However, effect of IPE field on k of the SL has not been studied. In this article, we have explored effect of IPE field on k of GaN/In\textsubscript{x}Ga\textsubscript{1-x}N/GaN SL for indium content ≤ 30%.
2. Theoretical Model

2.1 IPE field and its effect
Figure 1 shows structure of a GaN/InGaN/GaN SL. Total polarization in a layer is \( P = P^{\text{ph}} + P^{\text{el}} \). In this SL, GaN is barrier while InGaN performs as well. The interfacial electric field due to total polarization in a well \( E_w \) (or in barrier \( E_b \)) is \[ E_w = l_b (P_b - P_w) / (\epsilon_b l_w + \epsilon_w l_b) \]

\[ E_b = l_w (P_w - P_b) / (\epsilon_w l_w + \epsilon_b l_b) \]  
Here \( l_w, P_w(P_b) \) and \( \epsilon_w, \epsilon_b \) are width, polarization and dielectric constant of well (barrier) material, respectively. The IPE field is \( \sim 1 \) MV/cm.

Figure 1. Structure of GaN/InGaN/GaN SL.

Nitrides are piezoelectric semiconductors; so, as per Heckmann principle, the coupling between the strain field and electric field will induce additional electric polarization. This will contribute to elastic constant of the alloy. The elastic constant of In\(_x\)Ga\(_{1-x}\)N in the presence of IPE field has been found as [8]

\[ C_{44}^{\text{InGaN}} = C_{44}^{\text{InGaN}} + \left( \frac{\epsilon_{15}^2 + \epsilon_{31}^2 + \epsilon_{33}^2 + p_{3p}^2}{\epsilon_0} \right) \]  

where \( C_{44}^{\text{InGaN}} = 48 \times (1 - x) - 16 \times (1 - x) \) and \( C_{33}^{\text{InGaN}} = (224x - 398x1) \) GPa are elastic constant of In\(_x\)Ga\(_{1-x}\)N in the absence of IPE field [8]. The transverse and longitudinal phonon velocities are determined by \[ v_T = \left[ \frac{C_{44}^{\text{InGaN}}}{\rho} \right]^{1/2} \text{ and } v_L = \left[ \frac{C_{33}^{\text{InGaN}}}{\rho} \right]^{1/2} \] respectively; where \( \rho \) is material density. The average phonon velocity is obtained as \[ v = \sqrt{\frac{1}{3} (v_T + 1/v_T, 1 + v_L, 1/v_L)^{-1}} \]  

Debye frequency is determined by the expression \( \omega_d = \sqrt{3N/4\pi V_0} \) where \( N \) is the number of atoms per unit volume \( V_0 \). Debye temperature \( \theta_d \) is calculated by \( \theta_d = \hbar \omega_d / k_B \); where \( \hbar \) is Planck’s constant and \( k_B \) is Boltzmann’s constant.

2.2 \( k \) of GaN/InGaN/GaN SLs
The \( k \) of a semiconductor is written as sum of phonon and electron contribution \( k = k_{ph} + k_e \). It is demonstrated that \( k_e = 10^{-3} k_{ph} \). Thus, electronic contribution is neglected and for a semiconductor \( k = k_{ph} \). In solids, the thermal energy is carried by acoustic phonons. These phonons suffer scatterings due to other phonons, defects and interfaces. Here, normal scattering (\( \tau_n^{-1} \)), phonon-phonon Umklapp (\( \tau_u^{-1} \)), alloy or point-defect (\( \tau_p^{-1} \)), dislocation (\( \tau_d^{-1} \)), phonon-electron (\( \tau_{e-p}^{-1} \)) and boundary (\( \tau_b^{-1} \)) scattering rates are taken into calculation. The combined phonon scattering can be written as \( \tau^{-1} = [\tau_n^{-1} + \tau_u^{-1} + \tau_p^{-1} + \tau_d^{-1} + \tau_{e-p}^{-1} + \tau_b^{-1}] \)  

Thermal conductivity \( k \) of a layer of the SL is determined by Callaway expression (\( y = \hbar \omega / k_B T \)) [8]
\[ k = k_{ph} = \left[ \frac{k_B^4 T^3}{2\pi^2 h^3} \int_0^{\theta_p/\ell} \frac{e^\gamma}{(e^\gamma - 1)^2} dy \right] \]

In order to determine \( k \) of a SL, effect of interfaces has to be included. Interfaces generate thermal boundary resistance (TBR) which is computed by acoustic mismatch model (AMM) and diffuse mismatch model (DMM). If the interface is specular, then phonon transmissivity from a layer \( i \) to layer \( j \), \( \tau_s^{ij} \) is

\[ \tau_s^{ij} = \left( 4 Z_i Z_j \right) / \left( Z_i + Z_j \right)^2 \]

Where \( Z_i = v_i \rho_i \) is acoustic impedance, \( v_i \) the phonon group velocity and \( \rho_i \) the material density of the \( i^{th} \) layer. When interface is diffuse, diffuse transmissivity \( \tau_d^{ij} \) is

\[ \tau_d^{ij} = \left( C_i v_i \right) / \left( C_i v_i + C_j v_j \right) \]

Here \( C_i \) is specific heat of \( i^{th} \) layer. With \( (\mu_i = \cos \theta_i) \), the total transmission coefficient \( \Gamma_{ij} \) from material \( i \) to material \( j \) is

\[ \Gamma_{ij} = p \Gamma_s^{ij} + (1-p) \Gamma_d^{ij} = p \int \tau_s^{ij} \mu_i d\mu_i + (1-p) \int \tau_d^{ij} \mu_i d\mu_i \]

Using \( \Gamma_{ij} \) determined from Eq.(5), we can estimate TBRs from material \( i \) to material \( j \) by

\[ R_{ij} = \left( \frac{4\pi^2 h^3 v_i^2}{k_B^4 \Gamma_{ij} T_i} \right) / \left[ \int_0^{\theta_p/\ell} \frac{e^\gamma}{(e^\gamma - 1)^2} dy \right] \]

Normally, \( k \) of SL is a tensor because of different materials and interfaces which generates inhomogeneity in thermal flow. Depending on the direction of imposed temperature gradients, \( k \) is divided into two parts: thermal flow parallel to a layer plane is termed as in-plane thermal conductivity \( (k_{ip}) \); whereas thermal flow across the layers is cross-plane thermal conductivity \( (k_{cp}) \). The \( k_{cp} \) of GaN/In$_{0.1}$Ga$_{0.9}$N/GaN SLs with two layers per period is estimated by

\[ k_{cp} = \frac{l_1 + l_2}{k_1 + k_2 + 2R} \]

Here \( l_1 \) and \( k_1 \) are layer thickness and thermal conductivities of GaN(InGaN), respectively. \( R \) represents the TBRs between GaN and InGaN.

3. Result and Discussion

3.1 \( k \) of GaN/In$_{0.1}$Ga$_{0.9}$N/GaN SL

Figure 2 shows \( k_{cp} \) as a function of temperature for \( x=0.1 \) in the absence and presence of IPE field. The \( k_{cp} \) has linear dependence on temperature from 20 < \( T \) < 120 K, whereas from 120 < \( T \) < 250K \( k_{cp} \) has nonlinear dependence on temperature and \( k_{cp} \) is relatively independent of temperature for \( T \geq 250 \) K [3].
Figure 2. $k_{CP}$ of GaN/In$_0.1$Ga$_{0.9}$N/GaN SL as a function of temperature.

From figure 2, it can be found that $k_{cp}$ in presence of IPE field is lower than $k_{cp}$ in the absence of IPE field. The RT $k_{cp}$ of GaN (10nm)/In$_0.1$Ga$_{0.9}$N(5nm)/GaN SL in the absence and presence of IPE field is 5.720and 4.652Wm$^{-1}$K$^{-1}$respectively. This implies that IPE field reduces RT $k_{cp}$ and the reason for decrease of $k_{cp}$ is the increased TBR and decreased $k$ of layers due to IPE field. IPE field enhances phonon velocity and Debye temperature of the constituent material of the SL. Alloy effect raises scattering of high frequency phonons whereas boundary/interfaces enhance scattering of low frequency phonons. Low frequency phonons significantly contribute to $k$ of SL. IPE field increases the strength of boundary/interfaces scattering in the SL resulting in enhanced scattering of low and mid frequency phonons further. Phonon-mode mismatch between layers, which scales with the lattice mismatch at the interface is significantly modifying the $k$. Tong et al.[10] experimentally determined $k$ of In$_0.1$Gd$_{0.9}$N alloy as 13Wm$^{-1}$K$^{-1}$ with 233 nm thicknesses. Our predicted value is well below this alloy limit. Xu et al.[5] observed $k_{cp}$ of GaN/In$_0.1$ Gd$_{0.9}$N/GaN nanoporous SL as 4.2 Wm$^{-1}$K$^{-1}$which they attributed to additional phonon scattering from porosity of the sample. Zhang et al. [4] determined $k_{cp}$ of GaN/In$_0.1$ Gd$_{0.9}$N/ SL as 4.8 Wm$^{-1}$K$^{-1}$ which is closure to our predicted value.

3.2 $k$ of GaN/In$_{0.3}$Gd$_{0.7}$N/GaN SL

Figure 3 shows $k_{cp}$ of GaN/In$_{0.3}$Gd$_{0.7}$N/GaN SL as a function of temperature in the absence and presence of IPE field. Here also $k_{cp}$ curve can be divided into 3 regions: (i) linear dependence on temperature from 20 < T < 120 K which is attributed to coherent phonon transport through the layers and $T^3$ behaviour; (ii) nonlinear dependence on temperature from 120 < T < 250K which is due to the increased influence of incoherent effects and (iii) relatively independent of temperature for T > 250 K; but decreases very slowly with increasing temperature for T > 300 K [3-5].
Figure 3. $k_{cp}$ of GaN/In$_{0.3}$Ga$_{0.7}$N/GaN SL as a function of temperature.

The nature of $k_{cp}$ in these regions can be explained in a way similar to GaN/In$_{0.1}$Ga$_{0.9}$N/GaN SL under the fact that the low-frequency phonons are not affected in the temperature region $T < 120$K, whereas the high-frequency phonons are greatly affected for elevated temperature $T > 300$K due to anharmonic (Umklap) scattering which makes $k_{cp}$ to saturate with $T$. Our result shows that room temperature $k_{cp}$ of GaN/In$_{0.3}$Ga$_{0.7}$N/GaN SL in the absence and presence of IPE field is 5.221 and 4.282 Wm$^{-1}$K$^{-1}$ respectively. This implies that IPE field reduces room temperature $k_{cp}$ and is due to enhanced TBR under influence of IPE field. From figure 3, it can be seen that a minute decrease of $k$ is found for $T > 300$K due to increase of Umklap scattering. At room temperature authors such as Huxtable et al. [3], Tong et al. [10] and Saha et al. [11] have experimentally determined $k_{cp}$ as 4.6, 2.9 and 3.3 respectively for Si$_{0.6}$Ge$_{0.4}$/Si$_{0.7}$Ge$_{0.3}$ SL, In$_{0.3}$Ga$_{0.7}$N alloy and TiN/Al$_{0.7}$Sc$_{0.3}$N SL. Low $k$ in the alloy samples reveals presence of high amount of dislocations, impurities and inhomogeneities.

4. Conclusion
In this work, effect of interfacial polarization electric (IPE) field on cross-plane thermal conductivity $k_{cp}$ of GaN/In$_{x}$Ga$_{1-x}$N/GaN SL has been explored for Indium content $x \leq 0.3$. Our result reveals that TBR is enhanced at interfaces under the action of IPE field leading to decreased phonon transmission and more mismatches of acoustic properties. This caused reduction of $k_{cp}$ of the SL. TBR is found to be (2.10 to 5.30)$\times 10^{-9}$ m$^2$KW$^{-1}$ which is closer to the reported value of similar SLs. Room temperature (RT) $k_{cp}$ in the presence (absence) of IPE field for GaN (10nm)/In$_{x}$Ga$_{1-x}$N (5nm) SL are 4.652 (5.720) and 4.282 (5.221) Wm$^{-1}$K$^{-1}$ which demonstrate more than 20% reduction and are closely agreeing with experimental $k_{cp}$ of similar SLs. This work shows that desired value of $k$ can be achieved by tailoring electric field of nitride SL for maximum power production.

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