A comprehensive simulation model based on the Boltzmann transport equation and the Callaway model for thermal conductivity was presented in the article to calculate the thermoelectric characteristics of both Al$_x$Ga$_{1-x}$As and Ga$_x$In$_{1-x}$P materials. The details of our computational approach for Al$_x$Ga$_{1-x}$As can be found in ref. [2]. Here we describe our attempts and results to model the thermoelectric properties of Ga$_x$In$_{1-x}$P. We optimized our model by fitting the empirical data of Hall electron mobility versus temperature and carrier concentration [3,4,5] as well as thermal conductivity versus both composition and temperature [6,7,8,9]. The resulted set of material parameters were used for both electrical and thermal properties calculations. Therefore, the model parameters were validated based on the main transport quantities. The input data to the model calculations were the band structure parameters, doping concentration, the range of temperature, and the composition. All calculations were based on three conduction energy bands. The determined model parameters for Ga$_x$In$_{1-x}$P are given in Tables I and II.

The values of the band related properties are given in Table I. These parameters were kept constant in all the calculations versus composition, carrier concentration and temperature. The second sets of parameters which are lattice dependent and directly affect the lattice thermal conductivity are presented in Table II. The primary step in our calculations is proving the robust evidences about reliability of our model to accurately predict the thermoelectric properties of Ga$_x$In$_{1-x}$P system and to obtain a satisfactory fit of the experimental data. In order to show this reliability, we made an effort to fit the previously reported data for Ga$_x$In$_{1-x}$P material including Hall electron mobility versus composition, thermal conductivity versus temperature, and thermal conductivity versus composition.

Figure S1-(a, b) present the experimental and calculated electron mobility versus temperature (for different doping concentrations) and doping concentration (at different temperatures) for InP, GaP,
and Ga$_x$In$_{1-x}$P. Figure S1-(c) depicts the experimental and calculated thermal conductivity versus temperature for GaP and Ga$_x$In$_{1-x}$P. Figure S1-(d) illustrates the thermal conductivity of Ga$_x$In$_{1-x}$P versus x (Ga content). In all figures (except Figure S1-(d)), the experimental data are shown by symbols and the calculated results are displayed by solid lines. In Figure S1-(d), the symbols are Adachi’s model calculations of the thermal conductivity versus x and solid line shows our calculations [7,8]. A comparison between the experimental and calculated values in all figures show good agreements, which evidences the reliability of the predicted properties of Al$_x$Ga$_{1-x}$As and Ga$_x$In$_{1-x}$P.

Figure S1: (a, b) The experimental (symbols) and calculated electron mobility (solid lines) for InP, GaP, and Ga$_x$In$_{1-x}$P, respectively, versus temperature and doping concentration. (c, d) The experimental (symbols) and calculated (solid lines) lattice thermal conductivity of GaP and Ga$_x$In$_{1-x}$P versus temperature. (d) The calculated from ref [9, 10] (symbols) and our calculated (solid line) lattice thermal conductivity of Ga$_x$In$_{1-x}$P versus x.
Table I: Ga$_x$In$_{1-x}$P lattice parameters used to calculate the electrical conductivity

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Static dielectric constant</td>
<td>13.18-3.12$x$</td>
<td>[10]</td>
</tr>
<tr>
<td>High frequency dielectric constant</td>
<td>11.6-3.44$x$</td>
<td>[10]</td>
</tr>
<tr>
<td>Bulk modulus (N/m$^2$)</td>
<td>$(7.1+1.7)x\times10^9$</td>
<td>[9]</td>
</tr>
<tr>
<td>Mass density (kg/m$^3$)</td>
<td>4810-670x</td>
<td>[9]</td>
</tr>
<tr>
<td>Energy at $\Gamma$ point</td>
<td>1.418+0.77x+0.648x$^2$</td>
<td>[11]</td>
</tr>
<tr>
<td>Energy at L point</td>
<td>1.958-0.019x+0.688x$^2$</td>
<td>[11]</td>
</tr>
<tr>
<td>Energy at X point</td>
<td>2.369-0.152x+0.147x$^2$</td>
<td>[11]</td>
</tr>
<tr>
<td>Conduction band (CB) effective mass at $\Gamma$ for GaP</td>
<td>0.09</td>
<td>[9]</td>
</tr>
<tr>
<td>CB longitudinal effective mass at L for GaP</td>
<td>1.2</td>
<td>[9]</td>
</tr>
<tr>
<td>CB transverse effective mass at L for GaP</td>
<td>0.15</td>
<td>[9]</td>
</tr>
<tr>
<td>CB longitudinal effective mass at X for GaP</td>
<td>1.12</td>
<td>[9]</td>
</tr>
<tr>
<td>CB transverse effective mass at X for GaP</td>
<td>0.22</td>
<td>[9]</td>
</tr>
<tr>
<td>CB effective mass at $\Gamma$ for InP</td>
<td>0.08</td>
<td>[9]</td>
</tr>
<tr>
<td>CB longitudinal effective mass at L for InP</td>
<td>0.25</td>
<td>[9]</td>
</tr>
<tr>
<td>CB transverse effective mass at L for InP</td>
<td>0.25</td>
<td>[9]</td>
</tr>
<tr>
<td>CB longitudinal effective mass at X for InP</td>
<td>0.32</td>
<td>[9]</td>
</tr>
<tr>
<td>CB transverse effective mass at X for InP</td>
<td>0.32</td>
<td>[9]</td>
</tr>
<tr>
<td>Valence band (VB) effective mass (heavy hole)</td>
<td>0.60+0.19x</td>
<td>[9]</td>
</tr>
<tr>
<td>VB effective mass (light hole)</td>
<td>0.089+0.051x</td>
<td>[9]</td>
</tr>
<tr>
<td>CB acoustic phonon deformation potential (eV)</td>
<td>5.1+7.6x</td>
<td>[10]</td>
</tr>
<tr>
<td>VB acoustic phonon deformation potential (eV)</td>
<td>-5.1+17.8x</td>
<td>[10]</td>
</tr>
<tr>
<td>Strain parameter for point defect scattering</td>
<td>20</td>
<td>[7]</td>
</tr>
<tr>
<td>CB acoustic phonon deformation potential (eV)</td>
<td>5.1+7.6x</td>
<td>[10]</td>
</tr>
<tr>
<td>VB acoustic phonon deformation potential (eV)</td>
<td>-5.1+17.8x</td>
<td>[10]</td>
</tr>
<tr>
<td>CB non-parabolicity (eV$^{-1}$) for L, $\Gamma$, X</td>
<td>0.5, 0.5, 0.5</td>
<td>This work</td>
</tr>
<tr>
<td>VB non-parabolicity (eV$^{-1}$)</td>
<td>0.83</td>
<td>This work</td>
</tr>
</tbody>
</table>

Table II: Ga$_x$In$_{1-x}$P lattice parameters used to calculate the lattice thermal conductivity

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Debye temperature (K)</td>
<td>301+144x</td>
<td>[9]</td>
</tr>
<tr>
<td>Bulk modulus (N/m$^2$)</td>
<td>$(7.1+1.7)x\times10^9$</td>
<td>[9]</td>
</tr>
<tr>
<td>Mass density (kg/m$^3$)</td>
<td>4810-670x</td>
<td>[9]</td>
</tr>
<tr>
<td>CB acoustic phonon deformation potential (eV)</td>
<td>5.1+7.6x</td>
<td>[10]</td>
</tr>
<tr>
<td>VB acoustic phonon deformation potential (eV)</td>
<td>-5.1+17.8x</td>
<td>[10]</td>
</tr>
<tr>
<td>Strain parameter for point defect scattering</td>
<td>20</td>
<td>[7]</td>
</tr>
<tr>
<td>Grüneisen parameter</td>
<td>0.045+0.055x</td>
<td>This work</td>
</tr>
<tr>
<td>Higher order phonon scattering</td>
<td>3-0.4x</td>
<td>This work</td>
</tr>
<tr>
<td>Ratio of normal to Umklapp scattering</td>
<td>1.0</td>
<td>This work</td>
</tr>
</tbody>
</table>

I. Relaxation times

The scattering processes in a material are taken into account by relaxation times. Various approximations of the relaxation times can be found in the literature.$^{12,13}$ The relaxation times used here incorporate nonparabolicity of the band. The relaxation time for the ionized impurity scattering is given by following formula$^{12,13}$
\[ \tau_{ii}^{-1} = \left( \frac{\sqrt{2}e^4N\varepsilon_0^2}{\pi^2\hbar^2} \right) \left( E(1 + aE) \right)^{\frac{1}{2}} \left( 1 + 2aE \right) \left( \frac{1}{q_D^2(a_D^2 + 8m_dE(1+aE))} \right) \]  

(1)

where \( q_D = \sqrt{\frac{N_f\varepsilon_0^2}{\varepsilon K_B T_L}} \).

The acoustic phonon scattering rate was calculated based on the effective deformation potential approximation.\(^{14}\)

The scattering rates for equivalent and non-equivalent valleys for electron transfer from i to j valley are given, respectively, by the following relations:\(^{15}\)

\[ \tau_{iv}^{-1} = \frac{\langle Z_e^{-1} \rangle^2D_i^2D_j^2}{\sqrt{2}\pi\rho\omega_{ij}\hbar^3}E_f \frac{1}{2} \times \begin{cases} N & \text{absorption} \\ N + 1 & \text{emission} \end{cases} \]  

in which \( E_f = \begin{cases} E + \hbar\omega_{ij} & \text{absorption} \\ E - \hbar\omega_{ij} & \text{emission} \end{cases} \)

(2)

\[ \tau_{iv}^{-1} = \frac{Z_j\langle m \rangle^2D_i^2D_j^2}{\sqrt{2}\pi\rho\omega_{ij}\hbar^3}\left[ E_f(1 + \alpha_fE_f) \right] \left[ 1 + 2\alpha_fE_f \right] G_{ij}(E_i, E_f) \times \begin{cases} N & \text{absorption} \\ N + 1 & \text{emission} \end{cases} \]  

(3)

in which \( G_{ij} \) is defined as \( G_{ij}(E_i, E_f) = \frac{(1 + \alpha_iE_i)(1 + \alpha_fE_f)}{(1 + 2\alpha_iE_i)(1 + 2\alpha_fE_f)} \). \( Z_e, D_i, \omega_{ij}, \rho \) and \( E_f \) are the number of equivalent valleys, deformation potential, the phonon frequency which allows the intervalley scattering, density, and Fermi energy, respectively. In case of non-equivalent intervalley scattering, \( Z_e - 1 \) is replaced by \( Z_j \) which is the number of available final valleys for scattering.

The relaxation time \( \tau_{DP} \) for acoustic phonon scattering is:\(^{16}\)

\[ \tau_0^{-1} = \frac{\pi k_B T D_i^2}{\rho v_s^2 \hbar} D(E), \quad \tau_0^{-1} = \tau_0^{-1} \left[ 1 - \frac{aE}{1 + 2aE} \left( 1 - \frac{D_v}{D_A} \right) \right] - \frac{8aE(1 + aE)}{3 \left( 1 + aE \right)^2 \frac{D_v}{D_A}} \]  

(4)

where \( E \) is the energy relative to the band edge, \( \alpha \) is the nonparabolicity parameter, \( D(E) \) is the density of states, \( \rho \) is the density, \( v_s \) is the sound speed, \( D_A \) and \( D_v \) are the conduction-band and valence-band deformation potentials, respectively. The values of the charge carrier deformational potentials used are listed in Table I.

The selected materials both are polar materials; so, we also calculated the polar longitudinal optical phonon scattering rates using the following relation:

\[ \frac{1}{\tau} = \frac{4\pi\varepsilon_0 \hbar}{3e^2N_0} \left( \frac{1}{1/e_{\infty} - 1/e_0} \right) \sqrt{\frac{2\hbar}{m_{DOS} \omega_{op}}} \frac{x + \beta x^2}{1 + 2\beta x} \]  

(5)

Here \( x = E/k_B T \) is the reduced energy, and \( \beta = \alpha k_B T \) is the reduced non-parabolicity.

To obtain the total relaxation time, we applied the Matthiessen’s rule:\(^{12}\)
\[
\frac{1}{\tau} = \frac{1}{\tau_{il}} + \frac{1}{\tau_{iv}} + \frac{1}{\tau_{ac}}
\] (6)

II. The main thermoelectric properties

The formulas were used to calculate the electrical conductivity, Seebeck coefficient, and the electronic thermal conductivity as applied in a multiband model of Boltzmann transport theory. The electrical conductivity and Seebeck coefficient are given by:

\[
\sigma = \frac{e^2 (2m_{DOS}^* k_B T)^{3/2}}{m_{nx} \pi^2 h^3} \int_{x_f}^{\infty} \left( -\frac{\partial f_0}{\partial x} \right) \tau(x) \frac{(x + \beta x^2)^{3/2}}{1 + 2\beta x} dx
\] (7)

\[
S = -\frac{k_B}{e} \left( \int_{x_f}^{\infty} \frac{\left( -\frac{\partial f_0}{\partial x} \right) \tau(x)(x + \beta x^2)^{3/2}}{1 + 2\beta x} dx \right) - x_{Fermi}
\] (8)

in which \(-\frac{\partial f_0}{\partial E} = \frac{1}{ek_BT} \frac{1}{e^{(E-E_F)/k_BT} + 2 + e^{-(E-E_F)/k_BT}}\) is the derivative of Fermi-Dirac distribution function, \(x = E/k_BT\) is the reduced energy, \(\beta = ak_BT\) is the reduced non-parabolicity, and \(\tau\) is the total scattering time. \(m_{nx}\) is the conductivity effective mass.\(^1\) This procedure is repeated for each temperature, doping concentration, and different bands allowing all of the thermoelectric properties to be determined over the desired temperature and doping concentration ranges for each band. For the multiband transport model, the effective total electrical conductivity and Seebeck coefficient can be calculated as

\[
\sigma_{tot} = \sum_l \sigma_l , \quad S_{tot} = \sum_l \sigma_l S_l
\] (9)

where \(\sigma_l\) and \(S_l\) are the electrical conductivity and Seebeck coefficients, respectively, for each individual band. The contribution of the carriers of different bands to the thermal transport is calculated using the Widemann-Franz law\(^1\)

\[
\kappa_e = \sum_l L_l \sigma_l T
\] (10)

in which \(T\) is the absolute temperature, and \(L_l, \sigma_l\), are the Lorentz number, and the electrical conductivity, for each band. The Lorenz number is known to vary with the carrier concentration especially at low carrier concentrations, where \(L\) can be significantly lower than the free-electron value, so, we calculated \(L\) as function of carrier concentration using

\[
L_l = \left( \frac{k_B}{e} \right)^2 \begin{bmatrix} 2\beta^{3/2} & \frac{1}{\beta^{3/2}} & -\frac{1}{\beta^{3/2}} \\ \frac{1}{\beta^{3/2}} & 1 & \frac{1}{\beta^{3/2}} \\ \frac{1}{\beta^{3/2}} & \frac{1}{\beta^{3/2}} & 1 \end{bmatrix}
\] (11)
where we have

\[ \kappa_{n,m} = \int_{0}^{\infty} \left(-\frac{\partial f}{\partial x}\right)x^n(x + \beta x^2)^m \tau(x)^k (1 + 2\beta x)^' dx \]  

and the summation is achieved over all contributing bands. The bipolar thermal conductivity can be calculated as

\[ \kappa_b = \frac{1}{2} T \sum_{i,j} \sigma_i \sigma_j (S_i - S_j)^2 \]  

where \( i, j \) are the valley indices, and \( S \) is the Seebeck coefficient.

### III. Thermal conductivity

The formalism introduced by Callaway\(^ {18} \) is used to calculate the lattice thermal conductivity:

\[ k_l = k_1 + k_2 \]  

where \( k_1 \) and \( k_2 \) can be expressed as:

\[ k_1 = CT^3 \int_{0}^{\Theta/T} \tau_C(x)G(x)dx \]  

\[ k_2 = CT^3 \beta I \]  

with

\[ \beta = \frac{\int_{0}^{\Theta/T} \tau_C(x)G(x)dx}{\int_{0}^{\Theta/T} \tau_N(x)G(x)dx} \]  

\[ I = \int_{0}^{\Theta/T} \frac{\tau_C(x)}{\tau_N(x)} G(x)dx \]

where

\[ G(x) = \frac{x^4 e^x}{(e^x - 1)^2}, \frac{1}{\tau_C(x)} = \frac{1}{\tau_N(x)} + \frac{1}{\tau_U(x)}, x = \frac{\hbar \omega}{k_B T}, m = \frac{\hbar}{k_B}, C = \frac{k_B m^3}{2\pi^2 v} \]

in which \( \hbar \) and \( k_B \) are Planck’s and Boltzmann constants, respectively. The phonon angular frequency, the phonon group velocity (sound velocity), Debye temperature and absolute temperature are indicated by \( \omega, v, \theta, \) and \( T, \) respectively. \( \tau_C, \tau_N \) and \( \tau_U \) represent combined, N (resistive), and Umklapp relaxation times, respectively. The ratio of the Umklapp to normal mode scattering was set to 0.2 in our model.

Total thermal conductivity is given by:

\[ \kappa = k_l + k_e + k_b \]  

where \( k_l \) represents the lattice part of thermal conductivity, \( k_e \) indicates the electronic part of it and \( k_b \) is the bipolar contribution to the total thermal conductivity.

The contribution of the charge carriers to the thermal conductivity is calculated considering different bands:\(^ {19} \)

\[ k_e = \sum_i L_i T \sigma_i \]  

(18)
which the summation is done over all the involved bands where:

\[
\lambda_{\alpha\beta}^{n\alpha m} = \int_{0}^{\infty} \left( -\frac{\partial f}{\partial x} \right) x^{\alpha}(x + \beta x^2)^{m} \tau(x)^{i}(1 + 2\beta x)^{j} dx
\]  

In the above mentioned equations \( k_B \) is the Boltzmann constant, \( x = E / k_B T \) is the reduced energy, \( \alpha \) and \( \beta = (k_B T)\alpha \) are the non-parabolicity parameters, \( \eta = E_f / k_B T \) is the reduced Fermi energy and \( f(x) = 1/(e^{(x-\eta)}+1) \) is the Fermi-Dirac distribution function. The factor \((1+2\beta x)\) is induced due to the energy dependency of the effective mass to the non-parabolic band.

The bipolar thermal conductivity is calculated according to:\(^{19}\)

\[
k_b = \left( \frac{k_B}{e} \right)^2 \frac{\sigma_i \sigma_2}{\sigma_i + \sigma_2} (S_1 - S_2)^2 T
\]  

where 1 and 2 indices refer to electrons and holes.

IV. Optimum doping concentration

For the case of Al\(_x\)Ga\(_{1-x}\)As, at \( x<0.26 \), \( zT \) vs doping concentration has two peaks, the first one is in the \( 10^{19} \) cm\(^{-3}\) range and second one is in \( 10^{20} \) cm\(^{-3}\) range. In this range of \( x \), the first \( zT \) peak is larger. At \( 0.26<x<0.4 \), \( zT \) vs doping concentration still has two peaks, but the second \( zT \) peak is larger. At \( x>0.4 \), \( zT \) vs doping concentration has only one peak. The optimum doping concentration for the two peaks are plotted in Figure S2S-a.

Figure S2: The optimum doping concentration versus composition for (a) Al\(_x\)Ga\(_{1-x}\)As and (b) Ga\(_x\)In\(_{1-x}\)P.

Also, for In\(_{1-x}\)Ga\(_x\)P, we calculated the optimum doping concentration for each \( x \) value, which is shown in Figure S2S-a.

V. Figure-of-merit versus composition
Figure S3 demonstrates the figure-of-merit versus composition for Al$_x$Ga$_{1-x}$As (panel (a)) and Ga$_x$In$_{1-x}$P (panel (b)). The $zT$ calculations for fixed doping values of $1.05 \times 10^{19}$ cm$^{-3}$ and $1.5 \times 10^{20}$ cm$^{-3}$, respectively, for AlGaAs and InGaP are also shown by dotted lines. In practice, it is difficult to reach doping in the range of high $10^{20}$ cm$^{-3}$ in AlGaAs. Therefore, the 2$^{nd}$ $zT$ peak for AlGaAs is difficult to reach. The calculated $zT$ for the 1$^{st}$ peak is very close to the $zT$ calculation assuming the fixed doping. Therefore, we reported the transport properties for a fixed practical doping concentration ($\sim 10^{19}$ cm$^{-3}$) for all the x values in the main manuscript.

It is evident that in GaInP the optimum doping curve has a negligible difference with the fixed doping curve. Therefore, the transport properties were reported for the fixed doping concentration of $1.5 \times 10^{20}$ cm$^{-3}$ in the main manuscript.

![Figure S3: Figure-of-merit versus composition for (a) Al$_x$Ga$_{1-x}$As and (b) Ga$_x$In$_{1-x}$P.](image)

[9] The data of GaP and InP were taken from [http://www.ioffe.ru/SVA/NSM/Semicond](http://www.ioffe.ru/SVA/NSM/Semicond). The data for Ga$_x$In$_{1-x}$P were estimated using the weighted average of the properties of GaP and InP.
[10] The data of GaP and InP were taken from SpringerMaterials: The Landolt-Bornstein online database [http://materials.springer.com/](http://materials.springer.com/). The data for Ga$_x$In$_{1-x}$P were estimated using the weighted average of the properties of GaP and InP.