Rapid Prediction of Phonon Density of States by Graph Neural Network and High-Throughput Screening of Candidate Substrates for Wide Bandgap Electronic Cooling

Mohammed Saif Ali Al-Fahdi

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RAPID PREDICTION OF PHONON DENSITY OF STATES BY GRAPH NEURAL NETWORKS AND HIGH-THROUGHPUT SCREENING OF CANDIDATE SUBSTRATES FOR WIDE BANDGAP ELECTRONIC COOLING

by

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ABSTRACT

Machine learning has demonstrated superior performance in predicting a vast range of materials properties. However, predicting a continuous material property such as phonon density of states (DOS) is more challenging for machine learning due to the inherent issues of data smoothing and sensitivity to peak positions. In this work, phonon DOS of 2,931 inorganic cubic structures with 63 unique elements from the Open Quantum Materials Database are calculated by high precision density functional theory (DFT). With these training data, we build an equivariant graph neural network (GNN) for total phonon DOS of crystalline materials that utilizes site positions and atomic species as input features. The computational cost of training the GNN model is several orders of magnitude cheaper than full DFT calculations. More interestingly, the trained GNN model can predict partial DOS of the constituent atomic species even if such data were not included in the training, which demonstrates GNN’s capability in predicting the species contributions (node-level) of partial DOS from the total DOS predictions without additional computational cost. We then deploy the trained GNN model to predict phonon DOS of 4,626 cubic materials with band gap >0.2 eV to search for thermally conductive substrates for cooling a few representative high electron mobility transistors (HEMT) in terms of high interfacial thermal conductance (ITC). Our results show that high vibrational similarity or phonon DOS overlap is not a necessary requirement to obtain high ITC as evidenced in BN/MgO interface with ITC of 1,044 MW/m²K despite of phonon DOS overlap of only 0.22. Moreover, we highlight that the LTC of substrates
does not always play a significant positive role in determining ITC when cooling HEMT devices. However, higher LTC substrates indeed implies a higher magnitude of heat flux that can be transferred from the interfacial region. This work demonstrates the power of GNN models and paves the way for high-throughput screening of novel crystalline materials with desirable high ITC for phonon-mediated thermal management of wide bandgap electronics.

**Keywords:** Graph Neural Network; Phonon Density of States; Interfacial Heat Transfer; High-Throughput Screening; Wide Bandgap Electronics; Thermal Management

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CHAPTER 1
INTRODUCTION

1.1. Thermal Management

The rapid progressive development of cheaper, lighter, smaller, and high-power density electronic devices nowadays had been a topic of interest for a long time. For this endeavor, scientists and engineers were pushed to bring about novel ideas of new design methodologies, materials, processes, and tools to accomplish such design requirements. Therefore, electronic devices have become extraordinarily faster and shrunk in size from microscale to nanoscale in the period of just two decades [1]. In fact, multiple devices run in a two-digit nanometers size, and next-generation node size of 14 nanometers or lower are targeted for next-generation technology electronic devices [1]. It has been reported that the reducing size and increasing the density of transistors along with other integrated circuit devices enhances the computing capabilities at the cost of decreasing power dissipation [2]. However, with such ambitious device requirements comes a great deal of issues such as combustion in Li-ion batteries [3], and device failures due to thermal management [4], thermal runaway [3], and electric current leakage [5]. The power required to operate some high-performance computing processors is around 250 W which leads to accumulating heat of approximately 1 kW [6]. Such heat can dissipate through some design methodologies such as heat pipes [7] and phase change materials [8]. However, that still does not change the fact that some devices need to combine multiple layers to fulfil various requirements that one material does not possess such as GaN that
leaks a large magnitude of current in high-electron-mobility transistor (HEMT) devices [5]. Therefore, studying the interfaces between two different material layers remains a topic of interest in the materials selection and design process for such devices.

1.2. Motivation for materials science

People have used materials for various uses in their lives since the dawn of time in all ages such as Silicon age, Steel age, Iron age, Bronze age and Stone age [9]. Nowadays, developing new materials with desirable properties and multi-design restrictions have become extremely crucial in several commercial and environmental applications for human beings. For example, electric cars require batteries with higher safety and a huge capacity for energy storage [10]. Since there is a high demand for safe power generation devices with smaller sizes, thermal management has become a critical topic and design restriction to prevent the failure of those devices [11].

The development and emphasis of producing new and novel materials for various technological applications had been a slow process performed by trial-and-error experiments throughout the past ages due to the lack of understanding in structure-property relationships. An extreme example of this prolonged experimental setup is the pitch drop experiment [12] which occurs when a single drop is formed, and the drop is formed approximately between 7 to 13 years. This trial-and-error methodology or framework of novel materials development was the setting stone and the first paradigm in the four paradigms [13] of materials science. Then, materials scientists gathered numerous experimental results and empirical knowledge such as the laws of thermodynamics emerged in the second paradigm. In the third paradigm, computational
methods such as Hartree-Fock and density functional theory (DFT) have emerged and been formulated in the 20th century accompanied and facilitated by the advancement of quantum mechanics, condensed matter physics, and computational power in computers which enabled material scientists to compute and predict material properties from first principles to a high level of accuracy when compared to experiments with exponentially less resources and computational cost. In the fourth and current paradigm, this data-driven paradigm has appeared due to the low cost of the computations in the third paradigm which motivated materials scientists to generate innumerable hypothetical materials which engendered the advent of multiple large materials datasets (more details in section 1.3.2. and Ref. [14-18]). In this paradigm, materials design has become even remarkably faster and computationally cheaper than the previous paradigms since the machine learning models usually take second to hours, whereas DFT calculations can take from a few hours up to a few days, and experiments can take between days to months. One other advantage in this paradigm is gaining more understanding and untangling more correlations in the structure-property relationships. In section 1.3.2., we will provide more details on how materials scientists utilized machine learning in predicting material properties and discovering novel materials for various applications.
1.3. Motivation for machine learning

Machine learning is a type of artificial intelligence (AI) and can be defined as algorithms that can learn from experiences and form explicit instructions based on those experiences. The model learns those inferences and draws patterns from the provided data through statistical methods. The model can then be implemented and utilized for predicting results from different data points that were not seen by the model before. Some examples of machine learning algorithms are linear regression, decision trees, gradient boosting, and neural networks. Linear regression is the simplest and probably the first algorithm that is usually taught in machine learning courses. Linear regression makes predictions from the simple linear formula shown below:

$$y_{pred} = w^T x + b$$  \hspace{1cm} (1.1)
where $y_{\text{pred}}$ is the predicted output from training the model based on the inputs $x$. The symbols $w$ and $b$ represent the weights and biases, respectively. $w$ and $b$ change in each iteration until $y_{\text{pred}}$ hopefully reaches the global minima (i.e., $y_{\text{pred}}$ with the lowest error).

Decision trees resemble flowcharts in the sense that nodes are tests on inputs, branches are the outcomes of those tests, and the leaf nodes represent the decisions (i.e., $y_{\text{pred}}$). Unlike linear regression, decision trees can actually train on nonlinear relations between the inputs and outputs. A tree like model which tends to yield even better results is gradient boosting. Extreme Gradient Boosting (XGBoost) has been developed in 2016 [19] after it was originally pioneered by Friedman in 2001 who named it “gradient boosting” [20]. Gradient Boosting was also named Gradient Boosted Regression Tree (GBRT) or Gradient Boosting Machine (GBM) [19]. Machine learning models that use the Gradient Boosting algorithm build a tree ensemble. In that tree ensemble, every tree makes predictions with regularized terms to avoid overfitting. Then, the final prediction from the algorithm is the result from summing over each prediction made by each tree.

The algorithm was utilized in winning multiple competitions at a website mainly used for data science named Kaggle in which many data scientists around the globe participate and demonstrate their coding skills in real-world datasets to win financial awards from those competitions [19]. Among 29 Kaggle competitions, Extreme Gradient Boosting was utilized in 17 scripts of the best performing models.

Neural networks are one of the most widely used machine learning algorithms. One of the simplest forms of neural networks is a feedforward neural network with two layers that can be represented by the following formula:

$$ y_{\text{pred}} = g(w^{(1)^T} + b^{(1)})w^{(2)^T} + b^{(2)} $$  \hspace{1cm} (1-2)
Where $g$ is the activation function such as relu (probably the most common one), sigmoid or any other type of activation functions. $w(#) \text{ and } b(#) \text{ are the learned weights and biases in the layers indicated by the number } #. \text{ Other neural networks can differ tremendously from equation (1-2) in which there could multiple layers and weight constraints. According to the “universal approximation theorem” [21], neural networks can predict and approximate any type of function to arbitrary accuracy even though changing the weights and biases in neural network may be extremely complicated and daunting at times.}

1.3.1 Machine learning applications in materials science

Machine learning has gained tremendous attention as a potent and robust approach to discover and explore functional materials in a broad materials space [22-26]. The success of machine learning is mainly due to its immense competence to correlating and predicting outputs provided with reasonable input features (descriptors) and sufficient high-quality data to detect hidden and uneasy-to-discover patterns from the provided data. Performing experiments to get high quality data of material properties can be extremely expensive in terms of cost, time, and resources. In contrast, high-throughput calculations via density functional theory (DFT) have largely been utilized to obtain a wide range of material properties with high precision and with no artificial input parameters [27-34]. Machine learning has been able to utilize atomic-level or compositional features along with structural features of various crystalline materials [35] and has demonstrated its excellence and massive success in predicting a wide variety of material properties with high accuracy comparable to DFT calculations including, but not limited to electronic properties such as refractive index [36], bandgap [37], and
superconductivity [38], mechanical properties such as bulk, shear, and Young’s moduli, Poisson’s ratio, and Vickers hardness [39-40], and thermal properties such as heat capacity [41], thermal conductivity [42], and Debye temperature [43]. Most material properties tackled in previous machine learning studies are discrete values and predicted based on the entire crystal structure, i.e., a specific crystalline topography corresponds to a single or several materials properties. However, some other material properties that are remarkably and equally significant to study are continuous curves and have spectral-like formatting [44], i.e., the materials properties as a function of dependent variables such as dielectric function, absorption coefficient, electron density of states (DOS), and phonon DOS. Continuous and spectral-like properties pose several inherent challenges for machine learning related to the data itself, such as the high-demanding requirements of the data smoothing process and the special attention to peak positions which might trick machine learning models in determining the appropriate weights [45-46].

1.3.2 Materials structure graph representation

In the recent years, graph neural networks (GNNs) [47-48] have attracted a good deal of attention due to their flexibility in modeling complex data structures in various fields such as social sciences [49], drug discovery [50], gene DNA [50], brain structures [51], and materials science [52]. GNNs are constructed to represent graph-like data structures and to establish machine learning models based on the constituent nodes with the node features and the connecting edges and links [53]. GNNs have already captivated the attention of researchers in the materials science community due to their capability in interpreting crystal structures constituted by atoms and bonds as graphs composed of
nodes and edges, respectively [53]. The rapid development of GNNs has been demonstrated in the growing number of GNN-based packages developed by materials scientists such as Crystal Graph Convolutional Neural Networks (CGCNN) [54], Atomistic Line Graph Neural Networks (ALIGNNN) [52], Global Attention Graph Neural Network (GATGNN) [55], and MatDeepLearn [56]. In this work, we implement a GNN model to predict total phonon DOS using only atomic positions and species as inputs. Interestingly enough, the GNN model is also capable of rendering partial phonon DOS from each specie (same node type) without explicitly training on them, which demonstrates GNN’s competence in accurately aggregating the partial DOS of each specie to accurately yield the total DOS. The general framework for the previously explained details is shown in Figure 1-2. The total phonon DOS predictions are then used to screen possible combinations of two candidate materials through direct contact with ultrahigh interfacial thermal conductance (ITC), which is beneficial for solving the critical issue of thermal management of high heat flux electronic devices such as HEMTs with wide bandgap materials [57-58].

Figure 1.2 Graph Neural Network model initial framework for total phonon DOS and partial phonon DOS predictions and novel HEMT interfaces discovery.
CHAPTER 2

COMPUTATIONAL DETAILS AND MACHINE LEARNING TRAINING

2.1 DFT Calculations

All the 2,931 cubic (i.e., space group number higher than 194) crystal structures used for training our GNN model are obtained from the Open Quantum Materials Database (OQMD) in crystallographic information file (CIF) format [27] which include binary, ternary, and quaternary compounds. These cubic structures are then re-optimized by performing first principles calculations with the converged computational parameters to reach the ground state using Vienna Ab Initio Simulation package (VASP) [59-61]. The convergence criteria for crystal structures optimization are $10^{-8}$ eV and $10^{-4}$ eV/Å for the total energy and atomic forces, respectively. The optimization calculations fully allow the cell shape, cell volume, and atomic positions to change to reach the global minimum of potential energy surface of each structure. The Pedrew-Burke-Emrzerhof (PBE) parametrization of the generalized-gradient-approximation (GGA) is applied to describe the exchange-correlation effects of electrons [62] within the projector augmented wave (PAW) pseudopotentials [63]. The kinetic energy cutoff of 520 eV is set for all materials. The Brillouin zone was sampled using the Monkhorst-Pack k-mesh depending on the lattice constants, with the density of k-points sampling of 0.2 Å$^{-1}$ [64] to guarantee high quality of DFT calculations. For each optimized structure, we generated 12 to 18 supercells, whose size depends on the symmetry of the structure (generally the total number of atoms in the supercells ranges between 80 and 200) with random displacement
of 0.03 Å for each atom in the supercell and evaluated the atomic forces for these
displaced supercells by VASP. The convergence criterion for supercell VASP runs is $10^{-6}$
eV and $10^{-4}$ eV/Å for the energy and atomic forces, respectively.

2.2 Phonons Calculations

Consequently, we fitted the harmonic (2\textsuperscript{nd} order) and anharmonic (3\textsuperscript{rd} order)
interatomic force constants (IFCs) by the compressive sensing lattice dynamics (CSLD)
method [65-67] with the 3\textsuperscript{rd}-order force constants truncated to the 3\textsuperscript{rd} nearest neighbor.
With the 2\textsuperscript{nd} order IFCs obtained, the phonon dispersions and phonon DOS are then
calculated using PHONOPY [68]. The quality of phonon dispersions and DOS of
selected structures is also validated by comparing with those obtained from the finite
displacement difference method. Using the 2\textsuperscript{nd} and 3\textsuperscript{rd} IFCs as inputs, the lattice thermal
conductivity (LTC) of cubic structures is calculated using the ShengBTE package [69].

2.3 AlmaBTE Calculations

The ITC for interfaces formed by two candidate materials in contact is calculated
using AlmaBTE package [70] which implements Monte Carlo simulations and uses
diffusive mismatch model (DMM) to approximate the interfacial phonon transmission
coefficient. High dense grids of $24 \times 24 \times 24$ and $15 \times 15 \times 15$ were implemented in the
calculations for cubic (such as cubic GaN (c-GaN)) and noncubic (such as wurtzite
hexagonal GaN (h-GaN)) structures, respectively. In total we filtered 4,626 cubic
materials with bandgap higher than 0.2 eV from the OQMD database for GNN model
prediction of total phonon DOS.
CHAPTER 3
RESULTS AND DISCUSSION

3.1. GNN model training and prediction of total phonon DOS

The general outline of our GNN model training and prediction is shown in Figure 3.1. The crystal structures are first converted into graphs within a certain cutoff after implementing periodic boundary conditions. It is worth noting that, determining the cutoff radius \( R_{\text{max}} \) is extremely crucial in training the GNN model because the model needs to have an adequate graph with sufficient information from the neighboring environment for each node. However, having a large cutoff radius increases the number of neighbors, but it also makes the graph bigger and consequently increases the computational cost. A smaller \( R_{\text{max}} \) makes the graph smaller and reduces the computational cost, but the graph may not capture enough data from the neighboring environment for each node (i.e., atom). Herewith, \( R_{\text{max}} \) is determined by executing a statistical analysis which outputs the lattice constants \((a, b, c)\) and the number of samples (i.e., materials) which have a particular range of lattice constants. Evidently, all the lattice constants must be equal \((a = b = c)\) for each material because all our training and prediction structures are cubic. Such statistical analysis is extremely helpful in calculating an optimal value for \( R_{\text{max}} \). The histogram with the statistical analysis for the lattice constants is shown in Figure 3.2a). Correspondingly, the \( R_{\text{max}} \) value can be determined by calculating the average lattice constants. The average lattice parameters in
the following format \((a/b/c)\) are as follows: 4.44/4.44/4.44 Å. From the lattice constants averages and statistical analysis in Figure 3.2a), \(R_{\text{max}}\) between 4.5 Å and 5.5 Å should be sufficient to capture the neighboring environment for each node and not too high to increase the computational cost without significantly improving the predictions, and finally an \(R_{\text{max}}\) of 5 Å is selected in this study.

![Diagram](image)

Figure 3.1 Detailed framework for the graph convolutional networks model. (a) Crystal structures as graphs within certain cutoffs with periodic boundary conditions and adding the node features and edge attributes. (b) Graph Neural Network (GNN) layers namely gated and convolutional NN layers change the weights and biases in each node i.e., atom. (c) Outputting the predicted total phonon DOS of entire crystal structure with backpropagation to minimize the loss in total DOS. (d) Outputting the partial phonon DOS of constitutive elements without backpropagation or loss calculations.

The training, validation, and testing data split with respect to the entire dataset (i.e., 2,931 materials) is approximately 80%/10%/10%, respectively, i.e., the number of materials in training, validation, and testing sets are 2,328, 283, and 320, respectively.
Figure 3.2  a) A histogram of lattice constants \((a, b, c)\) for all the training materials. b) A histogram of the number of neighbors with the selected \(R_{\text{max}} = 5\ \text{Å}\) for all the samples used in this work. c) Normalized and balanced training, validation, and testing splitting for all 63 elements from the dataset which ensures the balance in the dataset.

Having implemented the periodic boundary conditions indicates that the number of neighbors varies with each cutoff radius \(R_{\text{max}}\), so the number of neighbors should be higher with higher cutoff radius, and vice versa. The average numbers of neighbors for (train/valid/test) are as follows: 29.7 / 28.8 / 31.5. Figure 3.2b) shows the number of materials with ranges of the number of neighbors. Most of the materials have around 30 neighbors which is close to the averages shown above. Some anomalies with extremely
high number of neighbors such as 60 neighboring atoms do exist in the dataset and those materials must have high density. On the other hand, materials with low neighboring sites such as neighboring 15 also exist in the dataset which reveals the low density in those materials.

The balance in training, validation, and testing splitting is dictated by the elements to ensure that the GNN model trains total phonon DOS using all types of elements. The mass of an element is a known quantity for controlling the phonon DOS. Generally speaking, the phonon DOS for light elements tend to have higher frequencies whereas that of heavier elements tend to occupy lower frequencies, whereas heavier elements tend to result in lower frequency modes [43, 71-72]. Balancing the dataset is necessary prior to training machine learning models in general to avoid biased dataset so that successful training on all possible scenarios can be ensured. Figure 3.2c) shows the normalized and balanced training, validation, and testing splitting among all elements. The columns that represent a species are normalized to unity due to the unbalanced number of samples that contain each species. However, as long as the GNN model trains, validates, and tests on each species, the dataset is balanced.

The batch size used in the training process is 3, i.e., 3 graphs or materials are trained at a time. A total number of 776 batches are trained because the number of training materials is 2,328, so each graph neural network (GNN) layer receives 3 graphs in each batch. The GNN layers shown in Figure 3.1 are constructed using Pytorch [73] and Pytorch Geometric [74] and e3nn [43]. The equivariance of the GNN model is
confirmed by e3nn [43] which is innately equivariant. Relu activation functions are used with each layer. Essentially, the weights in each layer from a previous layer get updated according to the previous layers. The layers update the embeddings of each node in each iteration with forward propagation which is explained in more detail by Kipf et al. [75]. Each node starts with a feature vector representation as shown in Figure 3.3a). The feature vector contains a one-hot encoder vector which includes atomic mass indexed by the atomic number. In crystal structures, the number of feature vectors in each graph by each atom depends on the neighboring atoms within a certain cutoff (i.e., $R_{\text{max}} = 5 \text{ Å}$ in this work). The loss criterion used in the model is the Mean Squared Error (MSE) at each iteration (i.e., epoch). The loss or MSE in the model is calculated based off the total DOS predicted by the model and compared with the “true” DFT calculated total DOS. After each total DOS prediction epoch, the loss is calculated, and the error backpropagates to update the embeddings in the GNN layers. The learning rate controls how much the embedding can change which was taken to be 0.005 with a weight decay of 0.05 to decrease the learning rate in each iteration and help reach the global minimum as the iterations increase. The Adam optimizer is used in the model as an adaptive learning rate optimization algorithm at each step. The early stopping for the model is 15 epochs i.e., the iterations should stop after 15 epochs from when the model starts overfitting after reaching the best model with the lowest validation loss.
Figure 3.3 a) Graph with feature vector at each node methodology scheme, b) Training and validation loss in each epoch up to 60 epochs, c) randomly selected materials in the testing set of total phonon DOS. The black color represents the “true” total phonon DOS of the materials calculated by DFT.

The best model exists at the 45th epoch which has the lowest validation MSE as shown in Figure 3.3b). The total number of training epochs is 60. The model started to overfit after reaching the lowest validation MSE at the 45th epoch (i.e., the best model), so the model had to execute an early stopping. The model training lasted about 43 seconds, which is several orders of magnitude cheaper than DFT calculations. Each crystal structure takes about two hours until it reaches the ground state. Then, around 10 jobs on an average run to compute phonon DOS of one material using finite displacement method in PHONOPY [68], and each job lasted about 2 hours. The testing total DOS model predictions are shown in Figure 3.3c). The black color curves represent DFT calculated total DOS. The cyan and blue colored curves are the predicted total DOS.
The total phonon DOS output by PHONOPY every 0.5 THz up to 30 THz so the length of output array should be 60 for all the materials. The total phonon DOS in the testing set materials consists of materials that the model had not seen before in the training or validation sets. It is seen from Figure 3.3c) that the predicted total DOS curves in the testing set are close to the DFT calculated DOS, and that proves the model’s accuracy in predicting the total DOS of materials that the model did not train on. The prediction of total DOS in the testing set materials seem to be precise enough to capture the DFT curves. MSE for the total phonon DOS testing set is calculated to be 0.024 which is slightly higher than the MSE of the validation set (i.e., 0.021). This is normal since the materials in the testing set were not being trained on. It is observed that materials with light elements such as Al in AlRu (1st row, 4th column in Figure 3.3c) tend to have phonon DOS at higher frequencies. On the other hand, materials with heavy elements such as both Ba and Te in BaTe (3rd row, 3rd column in Figure 3.3c) possess phonon DOS at lower frequencies. The model predicted the total DOS for those materials accurately. Therefore, the model can predict phonon DOS of materials that contain heavy and light elements alike. Noisy total DOS is more noticeable with higher number of atoms and species such as the perovskite KRuO3 (2nd row, 6th column in Figure 3.3c)), quaternary Heusler GaMnNbRu (3rd row, 6th column), and full Heusler CrHf2Os (2nd row, 2nd column) show more noise in the phonon DOS curves, whereas binary materials such as BrCs (1st row, 1st column) and BaTe (3rd row, 3rd column) feature less noise in the phonon DOS spectrum.
Figure 3.4 shows the GNN predicted total phonon DOS of randomly selected materials and their partial phonon DOS to the right of each total phonon DOS plot. It is worth pointing out that the GNN model did not train on, compute the loss, or backpropagate the partial DOS results. The model is trained only on the total phonon DOS, and all the loss calculations, backpropagation, node embeddings updates were only performed to get accurate total phonon DOS with minimal loss. The node contributions of the partial phonon DOS to the total phonon DOS were outputted from the best model as shown in Figure 3.3b). The partial phonon DOS results show the contributions of each node with the same species in the respective graph or material to the total DOS which is analogous to the partial phonon DOS contribution summed up to obtain the total phonon DOS in DFT calculations. The results exhibit high matching accuracy between DFT partial DOS curves for each species and the predicted node contributions (partial phonon DOS) of each species. This can only be done if the model properly and accurately aggregated the species contributions to the total phonon DOS when updating the embeddings during the training process. These results demonstrate the capability of the GNN model in accurately predicting partial phonon DOS just by training on the total phonon DOS without extra computational cost and separate training to make partial phonon DOS (i.e., node-level) predictions.
Figure 3.4 Randomly selected materials with GNN predicted total phonon DOS on the first column and partial phonon DOS of the constituent species to the right. The black lines are DFT results. Good agreement between the DFT results of total phonon DOS and GNN confirms the accuracy of the trained GNN model. The agreement between DFT results of partial phonon DOS and the GNN model demonstrate the capability of the GNN model in accurately predicting partial phonon DOS without extra computational cost and separate training.

3.2. Screening substrates for cooling high electron mobility transistors by phonon DOS overlap.

HEMTs are outstanding devices for high-frequency and high-power applications [76-78]. Hexagonal GaN (h-GaN) is one of the most common and crucial materials in HEMT applications due to its desirable attributes, such as medium to high LTC, high electron mobility, wide bandgap, and high breakdown electric field [57-58]. However, some of the issues that HEMT devices encounter are current leakage [79-80] and heat
dissipation limitations from the excessive heating which cause large increase of temperature in the device channel [57]. Electric current leakage is normally solved using materials with large bandgap or various materials combinations of metal-insulator-semiconductor HEMT [81]. The heat dissipation issues in high power electronics are often solved using high ITC between the layers to dissipate the excessive heat more effectively and thus reduce the temperature increase in the device channel. Several materials have recently emerged with potential applications in HEMT device channels along with h-GaN, such as AlN [82] and BN [83]. It was also reported that cubic GaN (c-GaN) was used in HEMTs [84-85]. Here we calculate ITC and analyze for several combinations of heat sources and heat sinks from previously unexplored materials, based on the phonon DOS predicted using the trained GNN model. The model is used to predict phonon DOS of 4,626 materials with bandgaps larger than 0.2 eV and energy above convex hull of 0.5 eV or less screened from the OQMD database [27] to discover high phonon DOS overlap with one of the heat source materials mentioned earlier. Here we consider the effect of total phonon DOS on ITC. Further ITC analysis demonstrates that our model can accurately predict phonon DOS of new structures, and the predicted total phonon DOS can further explain most of ITC results associated with HEMT devices.

Figure 3.5a) illustrates how heat flows from the channel in HEMT device to the substrate that acts as a heat source. ITC plays a significant role because if the ITC is low, heat will not dissipate at a sufficient rate which poses temperature limitations to HEMT and eventually degrades the performance of HEMT devices. Figure 3.5b) shows the schematic of a representative temperature profile of a HEMT interface and how to
calculate ITC using necessary outputs such as heat flux and temperature drop at the interface. Table 3.1 shows several materials with high LTC from the dataset used to train the phonon DOS. Two additional materials, i.e., h-GaN and diamond (denoted as \( C_{\text{diamond}} \)), are added to the analysis since they are common materials in HEMT applications. h-GaN has space group \( P6_3\text{mc} \) (space group number: 186) and it is useful to compare h-GaN to other candidate materials in our phonon DOS dataset. Furthermore, diamond is considered in this analysis because it is used in many HEMT devices as a heat sink or substrate [57-58].

![Diagram of heat flux and temperature profile](image)

**Figure 3.5** a) A schematic outline on how the heat dissipates from the HEMT device channel into the substrate (i.e., heat spreader). b) Overall schematic of representative temperature profile on how to calculate interfacial thermal conductance (ITC) from Monte Carlo simulation outputs using heat flux and temperature difference at the interface.

Materials with high LTC are selected because they possess weak phonon anharmonicity, and they are traditionally considered to be promising candidates as heat sinks. The main condition to having high ITC at the heat source/sink interfaces has long
been thought to have large overlap between the phonon DOS of the two materials [88-92]. The formula for phonon DOS overlap is defined as follows:

\[
\text{Phonon DOS Overlap} = \frac{2 \int_{0}^{\infty} D_{\text{overlap}}(\omega) d\omega}{\int_{0}^{\infty} D_{\text{heat source}}(\omega) d\omega + \int_{0}^{\infty} D_{\text{substrate}}(\omega) d\omega}
\]  (3.1)

where \(D(\omega)\) represents the phonon DOS of a material, and \(\omega\) is the phonon frequency.

With the obtained 2\(^{nd}\) and 3\(^{rd}\) order IFCs, we utilize almaBTE [70] to calculate the heat flux and temperature drop across the interface between the heat source and heat sink (substrate). AlmaBTE implements Monte Carlo models in calculating ITC at the interface when phonons dominate interfacial thermal transport [70]. ITC was calculated for the interface between two layers with 100 nm thickness for each layer. ITC results between the heat source materials (i.e., AlN, BN, c-GaN, and h-GaN) and the heat sink materials from Table 3.1 are shown in Tables A.1 – A.4 in Appendix A. Each table has 5 columns: (1) various heat source–heat sink combinations with one HEMT material for each table, (2) OQMD ID for the heat sink, (3) LTC for heat sink, (4) phonon DOS overlap between the heat source and heat sink calculated by Equation (3-1), and (5) ITC between heat source and heat sink. The tables present the results for all high LTC materials in Table 3.1 regardless of the phonon DOS overlap. Furthermore, Tables A.1 – A.4 in Appendix A give out the ITC for other substrates with lower LTC and possibly higher phonon DOS. The objective is to see how ITC varies with diverse LTC (i.e., high, moderate, or low LTC) when the phonon DOS overlap is also varying between the heat source and substrate.
Table 3.1 Candidate materials (in alphabetical order) with high LTC to be analyzed for HEMT devices. The structure ID and bandgap information is obtained from the OQMD database [27].

<table>
<thead>
<tr>
<th>OQMD ID</th>
<th>Formula</th>
<th>Bandgap (eV)</th>
<th>LTC (W/mK) at 300 K</th>
<th>Heat source/sink classification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1218324</td>
<td>AlN</td>
<td>3.458</td>
<td>272.93 (high)</td>
<td>Heat source or sink</td>
</tr>
<tr>
<td>8235</td>
<td>BAs</td>
<td>1.416</td>
<td>2358.3 (ultrahigh)</td>
<td>Heat sink</td>
</tr>
<tr>
<td>1218562</td>
<td>BN</td>
<td>4.795</td>
<td>839.28 (ultrahigh)</td>
<td>Heat source or sink</td>
</tr>
<tr>
<td>5664</td>
<td>BP</td>
<td>1.367</td>
<td>532.39 (high)</td>
<td>Heat sink</td>
</tr>
<tr>
<td>1218583</td>
<td>BSb</td>
<td>0.81</td>
<td>420.02 (high)</td>
<td>Heat sink</td>
</tr>
<tr>
<td>1215492</td>
<td>C\textsubscript{diamond}</td>
<td>4.4</td>
<td>2966 (ultrahigh) [86]</td>
<td>Heat sink</td>
</tr>
<tr>
<td>1222562</td>
<td>c-GaN</td>
<td>2.026</td>
<td>199.27 (high)</td>
<td>Heat source or sink</td>
</tr>
<tr>
<td>1472729</td>
<td>h-GaN</td>
<td>2.2</td>
<td>335.37 (in-plane) and 316.29 (out-of-plane) (high) [87]</td>
<td>Heat source or sink</td>
</tr>
<tr>
<td>4566</td>
<td>SiC</td>
<td>1.586</td>
<td>470.57 (high)</td>
<td>Heat sink</td>
</tr>
</tbody>
</table>

Figure 3.6 shows 3D visualization of the ITC results with AlN, BN, c-GaN, and h-GaN as heat source in HEMT. The corresponding substrate LTC, detailed ITC values, and phonon DOS overlap for all cases reported can be found in Tables A.1 – A.4 in Appendix A. The height of the bar in Figure 3.6 reflects the magnitude of ITC values, while the color bar represents the phonon DOS overlap between heat source and heat sink (substrate). The results are grouped into 4 islands, corresponding to AlN, BN, c-GaN, and h-GaN as labeled as heat source. Most of the heat sinks or substrates have medium to low LTC and only a few have ultrahigh LTC. The top two materials in terms of substrate LTC
are $C_{\text{diamond}}$ and BAs. The highest bar, i.e., the highest ITC of 3,542 MW/m$^2$K, occurs at BN-$C_{\text{diamond}}$ interface as denoted by the dark brown color, with corresponding highest phonon DOS overlap of around 0.6. Many other interface combinations possess ITC over 1,000 MW/m$^2$K and those are listed in detail in Tables A.1 – A.4 in Appendix A. Figure 3.6 also illustrates that, medium to high ITC does not always occur at two materials with high LTC. Indeed, we found many high ITC interfaces, but the two constitutive materials only have medium LTC, such as those in the left-bottom corner of Figure 3.6. For instance, h-GaN/BSb interface possesses a moderate ITC of 598 MW/m2K although their phonon DOS overlap is high (0.51) and LTC is also high for both materials (316.29 and 420.02 W/mK for h-GaN and BSb, respectively). This result also confirms that the high phonon DOS overlap, or vibrational similarity does not necessarily lead to high ITC.

It is well known that the phonon DOS overlaps affect how the heat fluxes travel across the interface between the heat source material and substrate [89-93]. Figure 3.7 displays the phonon DOS overlap of some selected combinations of heat source materials and substrates. The heat source/substrate pairs are selected based on high and low ITC values for each heat source material except for c-GaN which is not shown here because it has a substantially similar phonon DOS behavior to h-GaN. For brevity, we only show phonon DOS overlap for h-GaN as the heat source with other substrates. Figure 3.7 demonstrates the cases of high and low phonon DOS overlap for each heat source material, where the phonon DOS of heat source and substrate are represented by the light-blue and light-yellow colors, respectively, and their phonon DOS overlap is indicated by the gray area.
Figure 3.6 3D bar plot for all ITC results using AlN, BN, c-GaN, and h-GaN as heat source in HEMT. The height of the bar reflects the magnitude of ITC, while the color bar represents the phonon DOS overlap between heat source and heat sink (substrate). The corresponding substrate LTC, detailed ITC values, and phonon DOS overlap can be found in Tables A.1 – A.4 in Appendix A.

Figure 3.7a) exhibits high phonon DOS overlap of 0.56 at AlN/BP interface with ITC of 1,832 MW/m²K (Table A.1). Figure 3.7b) shows the opposite trend in phonon DOS overlap results with a phonon DOS overlap of only 0.07 which explains why AlN/BSb interface has a low ITC of 220.2 MW/m²K. The high phonon DOS overlap in BN/C\textsubscript{diamond} in Figure 3.7c) and the ultrahigh LTC in both BN and C\textsubscript{diamond} illustrates why it has the highest ITC in this work (3,542 MW/m²K). Figure 3.7d) reflects the low phonon DOS overlap in BN/MgO. Surprisingly, the BN/MgO interface has an ultrahigh ITC of 1,044
MW/m²K although MgO has a moderate LTC of 51.04 W/mK and relatively low phonon DOS overlap of 0.22, which proves the exception that the high phonon DOS overlap (match) is not a requirement to obtain a high ITC. A similar phenomenon was also found in a recent experiment [94]. h-GaN/LiF interface was found to have an extremely high ITC of 1,237 MW/m²K, mainly due to their high phonon DOS overlap of 0.51 shown in Figure 3.7e). The phonon DOS occupying the wide range of frequency interval in C\text{diamond}, compared to the phonon DOS in the lower frequency range in h-GaN, caused the phonon DOS matching area to be as low as 0.14 and finally medium ITC of 400 MW/m²K for h-GaN/C\text{diamond} interface.

Figure 3.8 shows how the spectral heat flux varies along the 200 nm distance of heat source and substrate thicknesses with respect to the phonon frequency. The colormap represents the heat flux which has various ranges depending on LTC of the materials, where the red and blue colors denote high and low heat flux, respectively. The heat flux is extremely high in the overlapping regions of phonon frequencies in AlN/BP as shown in Figure 3.8a). Such spectral heat flux is significantly important to interfacial thermal transport. The heat flux for AlN/BP interface is high in the frequencies between 4.8 to 11.2 THz which is exactly where phonon DOS of AlN and BP overlap as shown in Figure 3.7a). There are some other intervals in which phonon DOS in AlN and BP overlap such as 11.2 THz to 15 THz and 21 THz to 24 THz, but it seems that those intervals of phonon DOS overlap have high interfacial phonon scatterings, which is why the high heat flux is low or does not seem to exist in those phonon frequency intervals as shown in Figure 3.8a).
Figure 3.7 phonon DOS overlap between HEMT materials, i.e., AlN, BN, and h-GaN, with various substrates. a) AlN/BP, b) AlN/BSb, c) BN/C\textsubscript{diamond}, d) BN/MgO, e) h-GaN/LiF, and f) h-GaN/C\textsubscript{diamond}. The left panels represent high phonon DOS overlap, while the right panels represent low phonon DOS overlap. The phonon DOS of heat source and substrate are always represented by the light-blue and light-yellow colors, respectively, while the gray area represents the phonon DOS overlap between the heat source and substrate.
The AlN/BSb interface has a low phonon DOS overlap as shown in Table A.1 in Appendix A and Figure 3.7b), which is why there is mild red color (mild heat flux) in Figure 3.8b). The only visible red color exists between the intervals 3.2 – 6.4 THz which is precisely where the small phonon DOS overlap occurs in those two materials. The BN/C钻石 interface shows high heat flux in the interval 8 – 24 THz, as confirmed by their phonon DOS overlap in Figure 3.7c). Besides, their phonon DOS overlap is still significant at higher-frequency region from 24 to 37 THz, but high phonon scatterings seem to exist at those phonon frequency intervals. Figure 3.8d) illustrates the low spectral heat flux in BN/MgO which is caused by the low phonon DOS overlap area (i.e., 0.22) in Figure 3.7d). The mildly high spectral heat flux in BN/MgO appears more clearly in the frequency intervals 5 – 19 THz, which is also a region of phonon DOS overlap. There is no other phonon DOS overlap at higher frequencies in MgO with BN, and as a result the blue color (mild heat flux) dominates those frequency intervals. One more crucial observation on BN/MgO is the maximum heat flux (around 24 MW/m²) which is lower than that encountered in all previous interfaces. We believe this is caused by the lower LTC in MgO compared to all the previous materials. Even AlN/BSb interface has a higher heat flux transport at the interface than BN/MgO. That shows the importance of LTC in both materials (i.e., heat source and substrate) to transport higher heat flux at the interface. Figure 3.8e) manifests the high spectral heat flux of h-GaN/LiF interface, which occurs at 2 – 7 THz. The phonon DOS overlap exists in the phonon DOS of h-GaN/LiF in the intervals overlap of 1 – 10 THz and 15 – 19 THz as shown in Figure 3.7e). However, the high heat flux only occurs in the interval between 2 and 7 THz but does not occur in the second interval. This again is caused by the high phonon scatterings
that occur in the optical phonons in h-GaN and LiF. Even though ITC is high at the h-GaN/LiF interface (1,237 MW/m²K), the heat flux transport does not seem to be high (around 25 MW/m²), and that originates from the low LTC of LiF (27 W/mK). Mild heat flux occurs at h-GaN/C_{diamond} interface in Figure 3.8f) in the frequency interval 4.8 – 6.4 THz, where the phonon DOS overlaps in both materials as shown in Figure 3.7f). h-GaN/C_{diamond} has a high heat flux due to the high LTC that bother materials intrinsically possess. The heat flux computations shown in Figure 3.8 from the Monte Carlo simulations are consistent with the phonon DOS overlaps in Figure 3.7. Figure 3.8 also highlights the importance of high LTC of heat sinks or substrates in thermal management applications, as high LTC indicates high heat flux that can be transported across the interface and quickly dissipated away in the substrates. Such factual insights regarding LTC and ITC can help in the inverse design process of new and novel two materials that are in contact using modern artificial intelligence (AI)-based inverse design algorithms such as generative adversarial networks [95-96]. Such design processes can be implemented into diverse applications in which thermal management is a design consideration such as batteries [3, 97], buildings [7-8, 98-100], novel electronic devices [4-5], etc.
Figure 3.8 Spectral heat flux at the interfaces corresponding to the cases in Figure 3-8. a) AlN/BP, b) AlN/BSb, c) BN/C\textit{diamond}, d) BN/MgO, e) h-GaN/LiF, and f) h-GaN/C\textit{diamond}. The red and blue color indicate the high and low heat flux transported by the specific frequency phonon modes, respectively.
CHAPTER 4
CONCLUSION

To summarize, we constructed an equivariant graph neural network model and trained it on phonon DOS of 2,931 cubic crystal structures with 63 unique elements calculated by high precision DFT. The predicted phonon DOS results from our GNN model match the DFT calculations to a large extent, which demonstrates the capability of the model in predicting spectral-like materials properties. Interestingly, the model presented accurate predictions of partial phonon DOS of the constituting species, which can be attributed to the accurate aggregation for the embeddings in the graph nodes, i.e., materials species, which confirms the proper aggregations of total phonon DOS in the GNN layers, and the partial phonon DOS (node level) predictions were performed without additional computational cost. From computational speed point of view, training GNN model is several orders of magnitude computationally cheaper than the full DFT calculations. We further screened materials by phonon DOS overlap to discover new combinations of materials with ultrahigh interfacial thermal conductance for heat dissipation of HEMT devices. The trained GNN model was used to predict the total phonon DOS of 4,626 non-zero bandgap cubic materials from OQMD database. The phonon DOS predictions from the trained GNN model are used in explaining the ITC results in terms of phonon DOS overlap area between the two materials. More importantly, we reveal a few unexpected cases where ultrahigh ITC does not necessarily occur at interfaces with large phonon DOS match, for example, BN/MgO interface with
phonon DOS overlap of only 0.22 but ITC of 1,044 MW/m²K. This indicates that vibrational similarity or high phonon DOS overlap is not a necessary requirement to get high ITC. Our finding thus offers a new route for designing and screening novel heat dissipation materials for thermal management of wide bandgap HEMT electronics. Finally, we demonstrate that, although the LTC of substrates does not have deterministic effect on ITC, it affects the magnitude of total heat flux that can be transported across the interface, and therefore the traditional strategy of searching high LTC materials for heat dissipation is still valid.
REFERENCES


[83] Bechlaghem, F. Z.; Hamdoune, A. 3D Simulation and Optimization of Characteristics of Al0.1ga0.9n/mn0.2ga0.8n High Electron Mobility Transistor with b0.03ga0.97n Back-Barrier Layer. *2021*.


APPENDIX A

Table A.1 ITC results using AlN as heat source in HEMT.

<table>
<thead>
<tr>
<th>Heat source – heat sink</th>
<th>Substrate ID</th>
<th>Substrate LTC (W/mK)</th>
<th>Phonon DOS overlap</th>
<th>ITC (MW/m²K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlN – BAs</td>
<td>8235</td>
<td>2358.3</td>
<td>0.36</td>
<td>701.46</td>
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<tr>
<td>AlN – BN</td>
<td>1218562</td>
<td>839.28</td>
<td>0.27</td>
<td>1028.78</td>
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<td>AlN – BP</td>
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<td>532.39</td>
<td>0.56</td>
<td>1832.04</td>
</tr>
<tr>
<td>AlN – BSb</td>
<td>1218583</td>
<td>420.02</td>
<td>0.07</td>
<td>220.24</td>
</tr>
<tr>
<td>AlN – C\text{diamond}</td>
<td>1215492</td>
<td>2966</td>
<td>0.21</td>
<td>949.82</td>
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<tr>
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<td>1222562</td>
<td>199.27</td>
<td>0.30</td>
<td>613.62</td>
</tr>
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<td>AlN – h-GaN</td>
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<td>329.22</td>
<td>0.32</td>
<td>665.49</td>
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<td>AlN – SiC</td>
<td>4566</td>
<td>470.57</td>
<td>0.43</td>
<td>1601.62</td>
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<td>AlN – LiH</td>
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<td>24.10</td>
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<td>1667.60</td>
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<td>0.47</td>
<td>1767.51</td>
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<td>AlN – YN</td>
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<td>20.087</td>
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<td>AlN – GeC</td>
<td>22473</td>
<td>260.29</td>
<td>0.44</td>
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<td>AlN – LiF</td>
<td>327854</td>
<td>13.213</td>
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<td>AlN – CaO</td>
<td>5181</td>
<td>23.823</td>
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<td>1011954</td>
<td>18.611</td>
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<td>946.97</td>
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<tr>
<td>AlN – TiMnCoSi</td>
<td>703376</td>
<td>29.026</td>
<td>0.35</td>
<td>969.21</td>
</tr>
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Table A.2 ITC results using BN as heat source in HEMT.

<table>
<thead>
<tr>
<th>Heat source – heat sink</th>
<th>Substrate ID</th>
<th>Substrate LTC (W/mK)</th>
<th>Phonon DOS overlap</th>
<th>ITC (MW/m²K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BN – AlN</td>
<td>1218324</td>
<td>272.93</td>
<td>0.27</td>
<td>1063.34</td>
</tr>
<tr>
<td>BN – BAs</td>
<td>8235</td>
<td>2358.3</td>
<td>0.13</td>
<td>255.41</td>
</tr>
<tr>
<td>BN – BP</td>
<td>5664</td>
<td>532.39</td>
<td>0.22</td>
<td>889.34</td>
</tr>
<tr>
<td>BN – BSb</td>
<td>1218583</td>
<td>420.02</td>
<td>0.07</td>
<td>109.56</td>
</tr>
<tr>
<td>BN – C\text{diamond}</td>
<td>1215492</td>
<td>2966</td>
<td>0.57</td>
<td>3541.63</td>
</tr>
<tr>
<td>BN – c-GaN</td>
<td>1222562</td>
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<td>0.15</td>
<td>417.59</td>
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<tr>
<td>BN – h-GaN</td>
<td>1472729</td>
<td>329.22</td>
<td>0.18</td>
<td>434.04</td>
</tr>
<tr>
<td>BN – SiC</td>
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<td>470.57</td>
<td>0.28</td>
<td>1111.69</td>
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<td>BN – LiH</td>
<td>325865</td>
<td>24.10</td>
<td>0.40</td>
<td>1120.97</td>
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<tr>
<td>BN – NaH</td>
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<td>BN – BaLiH$_3$</td>
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Table A.3 ITC results using c-GaN as heat source in HEMT.

<table>
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<tr>
<th>Heat source – heat sink</th>
<th>Substrate ID</th>
<th>Substrate LTC (W/mK)</th>
<th>Phonon DOS overlap</th>
<th>ITC (MW/m²K)</th>
</tr>
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<tbody>
<tr>
<td>c-GaN – AlN</td>
<td>1218324</td>
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<td>c-GaN – BAs</td>
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<tr>
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<td>455.71</td>
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<tr>
<td>c-GaN – BSb</td>
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<td>420.02</td>
<td>0.48</td>
<td>628.48</td>
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<tr>
<td>c-GaN – C\text{\text{diamond}}</td>
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<td>c-GaN – SiC</td>
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<td>0.22</td>
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<tr>
<td>c-GaN – LiF</td>
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<td>c-GaN – ScN</td>
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<td>c-GaN – YN</td>
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<td>c-GaN – GeC</td>
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<td>0.48</td>
<td>908.53</td>
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<td>c-GaN – TiMnCoGe</td>
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<td>808.84</td>
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<td>c-GaN – MgS</td>
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Table A.4 ITC results using h-GaN as heat source in HEMT.

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<tr>
<th>Heat source – heat sink</th>
<th>Substrate ID</th>
<th>Substrate LTC (W/mK)</th>
<th>Phonon DOS overlap</th>
<th>ITC (MW/m²K)</th>
</tr>
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<tbody>
<tr>
<td>h-GaN – AlN</td>
<td>1218324</td>
<td>272.93</td>
<td>0.32</td>
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<td>h-GaN – BAs</td>
<td>8235</td>
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<td>h-GaN – BN</td>
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<tr>
<td>h-GaN – BSb</td>
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<td>1223434</td>
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<td>h-GaN – YN</td>
<td>6913</td>
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<td>906.59</td>
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<td>h-GaN – TiMnCoGe</td>
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