Effects of Temperature on the Transport properties of a Graphene and Hexagonal Boron Nitride Bilayer

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Since its discovery in 2004, graphene (G) has attracted significant interest in recent years due to its thermoelectric properties, such as its high carrier mobility and its high Seebeck coefficient, which have made it a material of interest for new nanodevices and applications.

However, some of its properties such as its "zero bandgap[1]" or its high thermal conductivity[2] have impeded further work with the material. In recent years, this has led to the development of several proposals that modify the materials properties to suit the needs of the application. One of these solutions has been to deposit Graphene layers on top of other substrates like hexagonal Boron Nitride (hBN)[1,2].

Experimental and computational results have shown that the formation G-hBN bilayers may have desirable effects, such as allowing a degree of tunability on its bandgap and its carrier mobility, which may influence the transport properties of the material such as their Seebeck coefficient, their Conductivity, and their Power Factor[2].

This proposal becomes the motivation to study transport coefficients of G-hBN bilayers, and in this work, we would like to see what their dependence on Temperature is. As an initial prediction for the results, it is expected that the Seebeck coefficient, thermal conductivity and power factor would increase with higher temperatures.

Procedure

To determine the T dependence of the transport properties of the G-hBN bilayer. Computational simulations were used to determine both the electronic structure of the materials and its transport properties. These computational resources were provided by nanoHUB.

Density Functional Theory

First, we performed DFT calculations with Local Density Approximation (LDA) on the DFT Material Properties Simulator. The simulation was defined with a Wavefunction Kinetic Energy cutoff of 40 Ry, a Charge Density cutoff of 160 Ry, and a SCF Convergence Criterion of 10^{-5} Ry to assure energy precision.

As seen on Figure 1. For the G-hBN bilayer, a single AA stacking pattern was used (i.e. each of the Carbon atoms in Graphene is directly aligned on top of a Boron or a Nitrogen Atom.) with an interplanar distance of 3.2 Å.

The band structure and the density of states for the G-hBN bilayer were determined. Additionally, thanks to the computer simulation, input data was prepared to be used for the Landauer transport simulation.

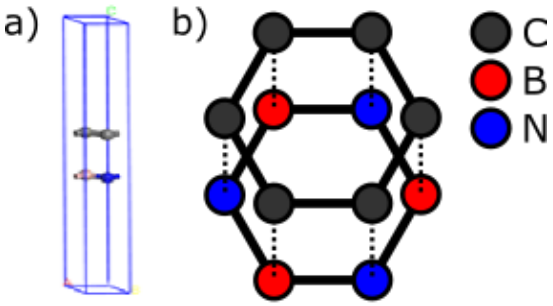

Figure 1: a) Unit crystal structure provided by collaborator for Calculations. b) Stacking pattern for G-hBN bilayer

Landauer Transport

The next step of this work was to use the results from the DFT calculations to calculate the thermoelectric properties of the G-hBN bilayer using LanTraP[3], a computer simulation based on the Landauer transport theory.

The simulation begins with determining the Distribution of Modes (DOM). This requires that the simulation is consistent with the input files obtained during DFT calculations, so they must include 30x30x1 k points and 16 Bands. The energy grid for this part of the simulation was between -20 to 20 eV with a dE of 0.001 eV.

With the DOM obtained, we proceed to obtain the Thermoelectric constants using a ballistic simulation, The Fermi energy grid was delimited between -16 eV to 16 eV, choosing a smaller interval than the energy grid for the DOM to maintain stability for the computations.

Results

DFT calculations

The resulting band structures and Density of states are shown on figures 2a and 2b respectively.

Starting with the energy bands, we expect from the literature that an interplanar distance of 3.2 Å would lead to a bandgap of ~0.2 eV.[2] In our calculations were able to confirm the presence of a band gap with a value of a similar magnitude 0.18 eV.

Additionally, running DFT calculations to determine the band gap of Graphene alone allows us to visually confirm some reliability in our calculations, as observed on Figures 3.a and 3. b.

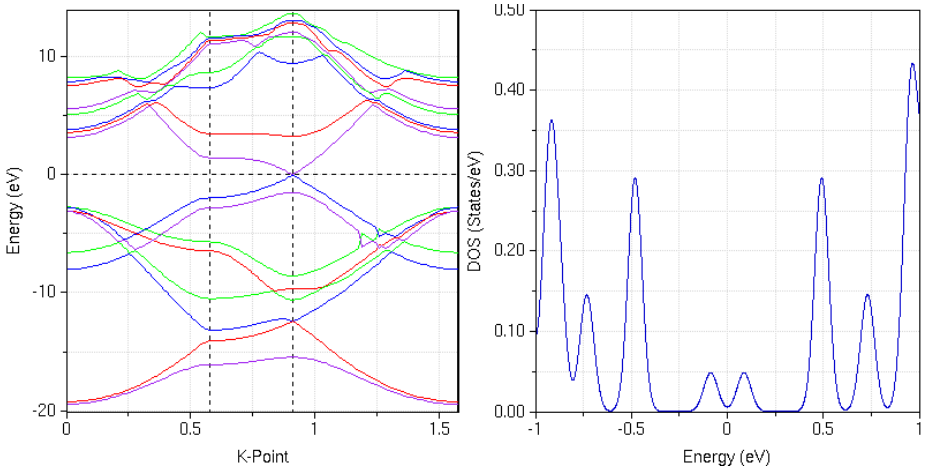

Figure 2: (a) G-hBN band structure along all symmetry points obtained from DFT calculations. (b) Density of states of G-hBN vs. energy near the band gap region. Initial unit cells were made by my collaborator.

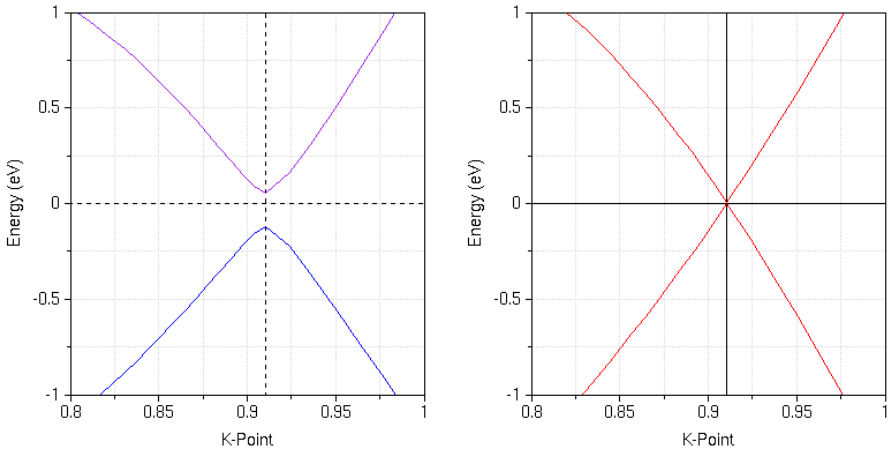

Figure 3: a) Zoomed bandgap for G-hBN. b) zoomed bandgap for Graphene

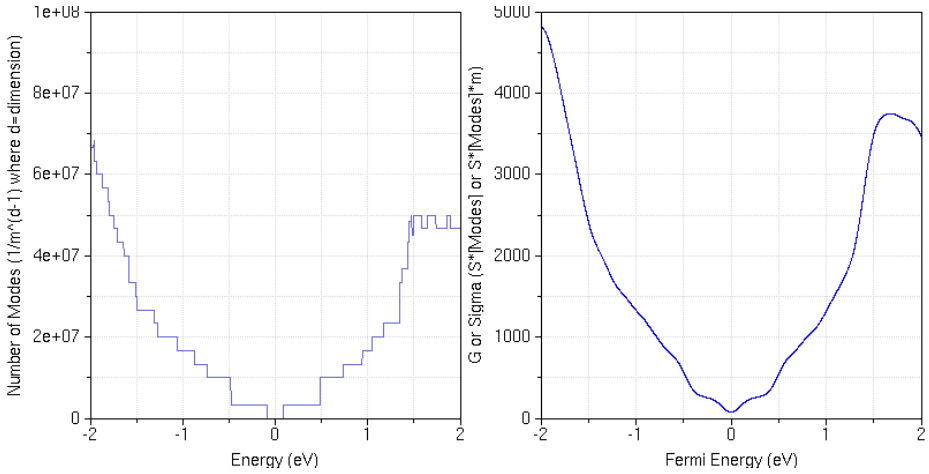

Figure 4: (a) Distribution of Modes vs. Fermi Energy for G-hBN near the band gap region. (b) Conductivity vs Fermi Energy

Landauer Transport

Results for the Landauer Transport Calculations can be observed on Figures 4 through 6. Also included are experimental and computational results from Duan, J. et. Al. for comparisons between the Seebeck Coefficient and the Power Factor for the G-hBN bilayer.[2]

As an initial observation, figures 5 through 7 confirm that there is a Temperature dependence for Thermoelectric constants such as the Seebeck Coefficient, the Power Factor, and the Thermal Conductivity. However, it is important to make comparisons between the calculations and actual experimental, results, since there are several discrepancies from the obtained results to the expected values.

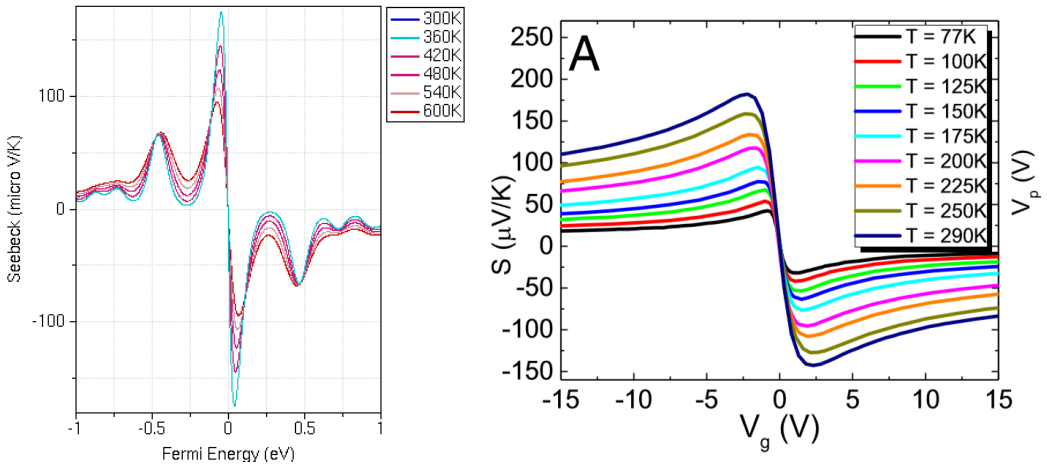

Figure 5: (a) Seebeck Coefficient vs. energy at different temperatures. (b) Measured Seebeck coefficient in a G-hBN device as a function of backgate voltage and temperature.

Starting with figure 5 to compare the Seebeck Coefficients between our calculations and experimental data. Two important trends can be observed that provide insights for our simulation. On one hand, similar coefficient values can be observed between 5.a, and 5.b and there is a similar trend in the transition from positive to negative values at 0. However, we can observe an inverse trend in that transition, as our calculations predict that S reduces with higher T's, while the experimental results shows an increase. Discrepancies between the results are also visible in Figure 6.a and 6.b. While the trend of values increasing with temperature can be observed, there is a higher discrepancy in values by a factor of ~10^4.

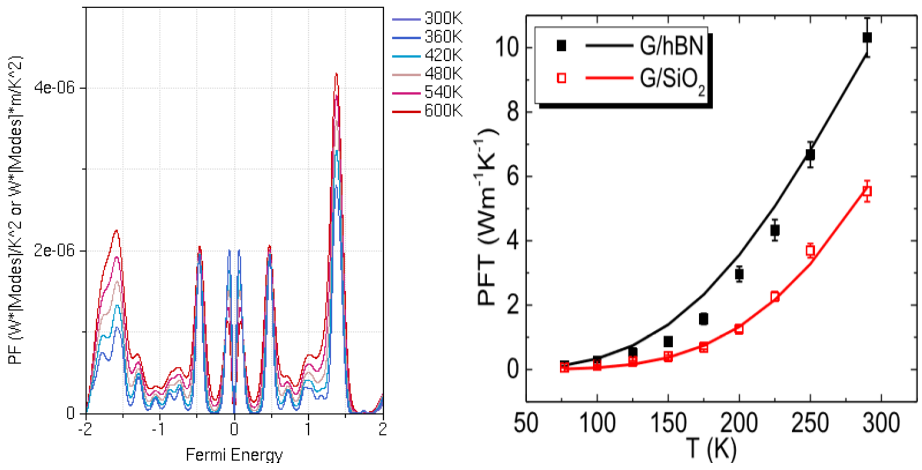

Figure 6. (a) Calculated Power Factor vs. Energy for G-hBN near the band gap region. (b) Measured PFT vs T values from two different Graphene devices with theoretical values (solid lines).

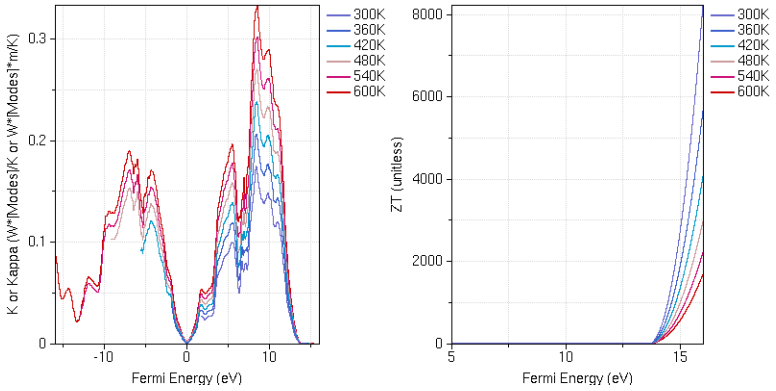

Figure: (a) Electron Thermal Conductivity vs. Fermi Energy at different temperatures. (b) Electronic ZT vs. Energy for G-hBN near the band gap region.

Discussion and Further Work

The discrepancies between the literature, experimental results, and our computations can be explained by several factors. On one hand, there is a need to improve on the DOM, which can be done by increasing the number of k points used during DFT calculations and increasing the step size for the original Energy.

These variables are also important because they are also related to the band-counting method used to determine the DOM. While this method provides faster calculations to other interpolation methods since it doesn't require group velocity, this method has a tendency of underestimating the DOM, which may result in a jagged curve as the one seen on Figure 4.a.[3]

By defining proper parameters that may allow a better DOM to be produced, further work on Landauer transport can be done to further inquire how do the thermoelectric properties of bilayer devices such as G-hBN get modified by interlayer distance and layer stacking.

Conclusions

Computer simulations were used to study the effects of Temperature on the Thermoelectric properties of a G-hBN bilayer. While the results show discrepancies in terms of value ranges and tendencies to experimental data and results from the literature, there's value in the calculations done to observe trends that may lead to new nanodevices and to further the study of 2D materials.

Acknowledgements.

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Bibliography

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