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Thermal Conductivity of Buckled and Puckered Arsenene: An Equilibrium Molecular Dynamics (EMD) Simulation

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Abstracts: This study aims to calculate the thermal conductivity of buckled and puckered arsene using Molecular Dynamics simulation through Large-scale Atomic/Molecular Massively Parallel Simulation (LAMMPS). Arsenene is the monolayer of arsenic, that belongs to one of its allotropes, the gray metallic arsenic. The temperature varied from 100K, 300K, 500K, and 700K with a fixed dimension of 10nm x 50nm. Also, the length of the material varied from 10nm, 20nm, and 30nm. 40nm and 50nm at a fixed temperature of 300 K with constant width of 10nm. Results show that the main contributor to the increase of thermal conductivity is due to the phonon means free path. As the length increases, the thermal conductivity also increases because of its long mean free path. Moreover, the decreasing thermal conductivity as to the increase in temperature is due to phonon mean free path shrinking, resulting in a reduction in thermal conductivity. At room temperature, the thermal conductivity of buckled and puckered arsenene is found to be 1.8 W/mK and 1.3 W/mK, respectively. This study will provide fresh insight into the thermal properties of buckled and puckered arsenene, which will be useful for nanoelectronics applications.

Keywords: LAMMPS, Thermal Conductivity, Buckled Arsenene, Puckered Arsenene, Molecular Dynamics, Equilibrium Molecular Dynamics.

1. INTRODUCTION

The discovery of graphene and its exceptional properties give a new type of class of material known as "2D materials". This discovery becomes an outlet for to research community to look for the most ideal material that possesses different kinds of properties. Thus, these 2D materials have a unique application in different fields of applications and that makes them special among the rest [1, 2]. 2D materials are categorized into a large family of materials. These include elemental substances [3, 4], transition metal dichalcogenides (TMDs) [5, 6], and group III-IV compound [7] and MXenes [8], etc. One of them is a class of elemental layer group-V materials (phosphorus, arsenic, antimony, and bismuth), that recently gained popularity due to its potential and broad vision in electronic applications. Compared to other layered elemental materials, it is found that it exhibits a relationship with layer number, temperature, or applied strain [9]. One of the elements under group V is arsenic, it has three common types of allotropes metal gray, yellow, and black As, where the most stable among them is gray metal As. The layered lies within the rhombohedral structure in a β phase also known as Buckled arsenene. By added heat of approximately 370K, the layered orthorhombic appears in an α phase known for its puckered form arsenene [10, 11]. This material possesses an indirect band and can be further tuned into direct by applying the method of biaxial strain [12]. To know the efficacy of a material, one must look for its thermal properties. This plays an important role, especially in the operation of mechanical and electronic devices which judge the efficacy of material on how often it conducts heat. There are many methods to determine it bases on molecular dynamics simulation. For instance, by doing the non-equilibrium dynamics, equilibrium molecular dynamics, first-principle calculation, and ab initio.

This paper aims to investigate the thermal conductivity of buckled and puckered arsenene using EMD simulation in the Green Kubo method. To determine the behavior of the materials in different temperatures and lengths. It will be important for nanoelectronics applications and as thermal management material.

2. COMPUTATIONAL DETAILS

This work used a Molecular dynamic (MD) simulation performed using LAMMPS [13]. The program allows receiving information of instructions from the input script that was made manually where the input parameters and other information were used to run a program. One most important thing to consider when doing an MD simulation is choosing a suitable interatomic forces/pseudopotential since this is dependent on accuracy calculations of force and potential energy of atoms in a simulation. The interatomic forces/pseudopotentials being used are Stillinger Weber Potentials [14], to describe the interactions of As atoms in a buckled and puckered structure. The main concept in creating and equilibration of the simulation cell is typical for EMD simulations which were later used to determine the thermal conductivity of a material in the Green Kubo method.

The first thing to do was construct a unit cell for both Arsenenes by knowing its lattice parameters and space group. Buckled arsenene belongs to the P3m1 space group with a height of h = 1.40 Å given by the lattice constant of 3.16 Å, bond length of 2.51 Å, and initial lattice parameter of a = 3.607 Å with space vacuum of c = 100.0 Å This is added along [001] miller directions [14, 15]. Puckered arsenene belongs to the Pmna space group with a lattice parameter of a = 3.677 Å, b = c = 4.765 Å, and has a value of the base for the rectangular unit cell of $a1$ = 4.77 Å, a2 = 3.68 Å and a3 = 11.11 Å. This consists of four As atoms in the basis unit cell [13, 16]. For both materials, the temperature was varied by 100K, 300K, 500K, and 700K with a fixed dimension of 50nmx10nm. Then, at room temperature 300K with corresponding different lengths of 10nm, 20nm, 30nm, 40nm, and 50nm at a fixed width of 10nm. Figure 3.1 shows an illustration for considered materials to be modified.

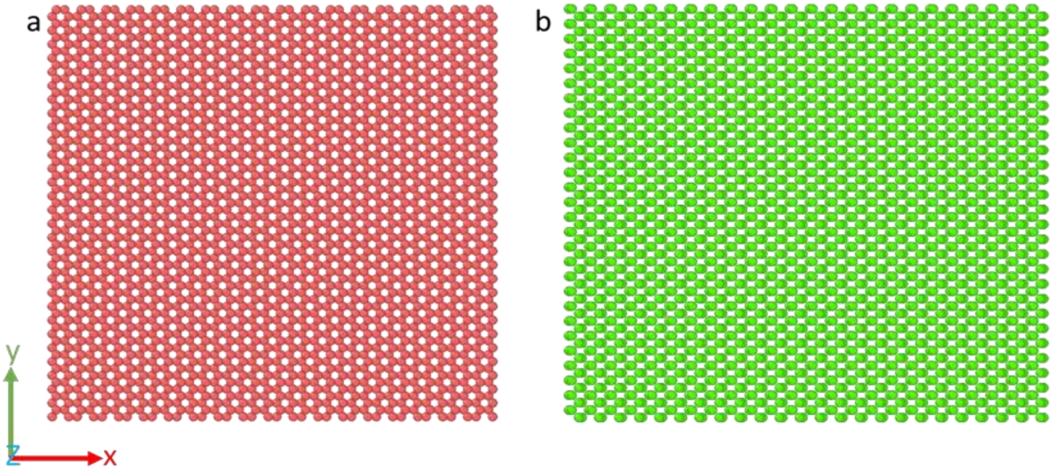

Figure 3.1. Sample dimension for 10nmx10nm for Buckled arsenene and b. Puckered arsenene the red and green represent As atoms.

After gathering all necessary data, the structure was minimized using the conjugate gradient (CG) algorithm following periodic boundary conditions with a timestep of 0.001 fs. For obtaining the steady states of the structure, the NPT command applies. During this phase, the velocity of the atoms was rescaled periodically, which allows the simulation to be thermostated. The effect of periodically rescale velocity is to inhibit the system from naturally evolving the influences of the formation of phonons[17, 18]. The data was then used to determine the thermal conductivity of the materials by the use of EMD simulation using the Green Kubo formula. This is derived from the fluctuation-dissipation theorem and linear response theory which relates the thermal conductivity and heat current autocorrelation function (HCAF)[19] as follows:

$$k=V/(3k_\beta T^2)\int \langle \vec{J}(0)\cdot\vec{J}(t)\rangle dt$$

where k is the thermal conductivity, V is the system volume, k_β is the Boltzmann constant, and $\langle \vec{J}(0)\cdot\vec{J}(t)\rangle$ is the heat current autocorrelation function.

3. RESULTS AND DISCUSSIONS

3.1. Length dependence of thermal conductivity

The thermal conductivity of buckled arsenene as the length is varied from 10nm, 20nm, 30nm, 40nm, and 50nm at a fixed width of 10nm at room temperature of 300K was shown in Figure 3.2. In a range of 10nm to 40nm, the results show that it somehow slowly increases then sudden increases from 40nm to 50nm. The increase of thermal conductivity as to the length is due to the participation of phonon interaction which create a heat transfer in a phonon scattering behavior. The longer length the more interactions happened; this is the reason why the thermal conductivity increases as to length increases.

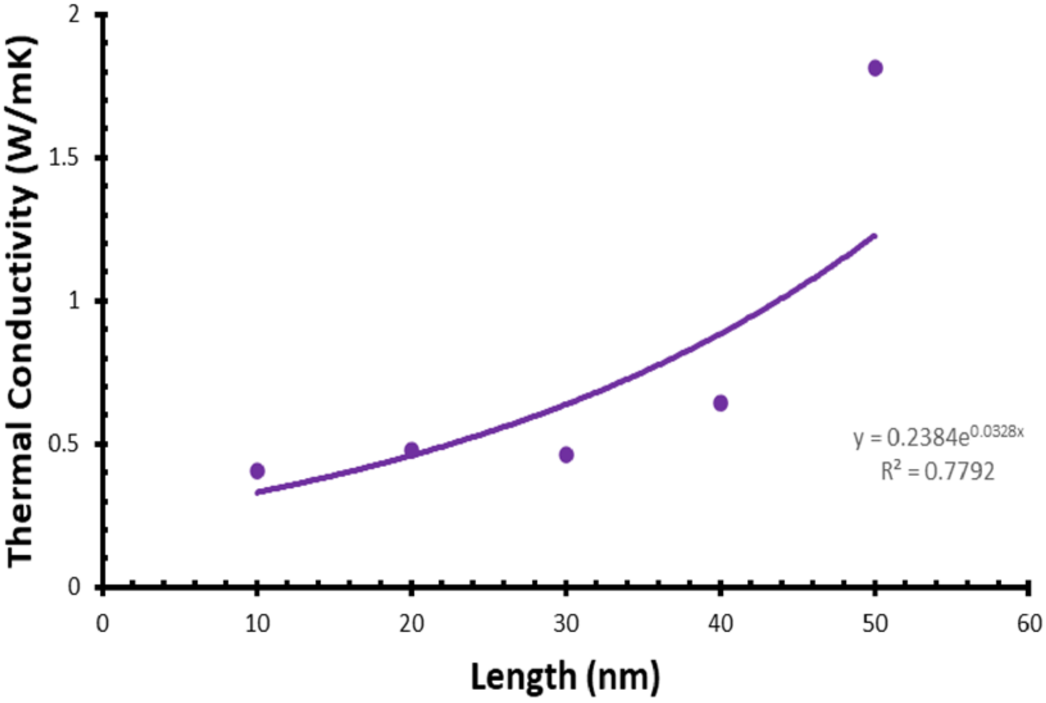

Figure 3.2. Thermal conductivity of Buckled arsenene as a function of length at room temperature (300K) at a fixed width of 10nm. The trend shows that the thermal conductivity at 10nm to 40nm thermal conductivity has a slow rate of increase. From 40nm to 50nm it shows a rapid increase.

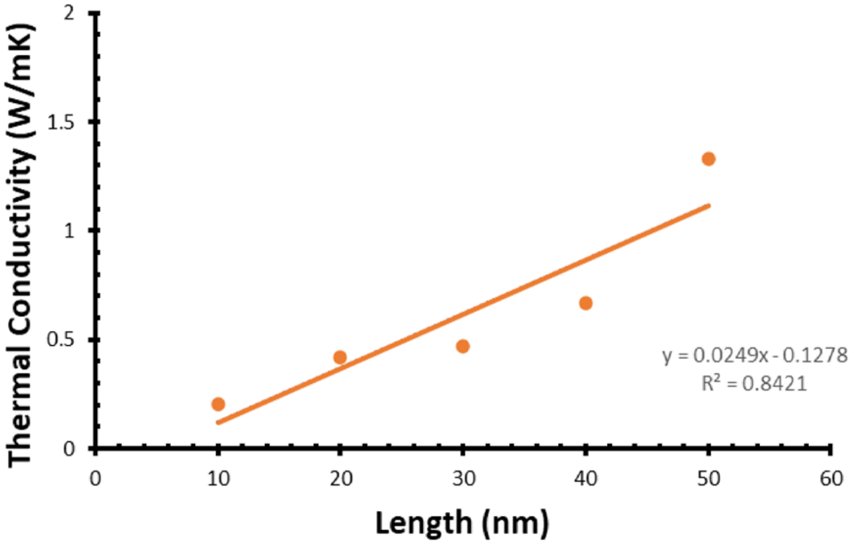

Figure 3.3. Thermal conductivity of Puckered arsenene as a function of

length at room temperature (300K) at a fixed width of 10nm. The trend shows a smooth increase in thermal conductivity as length increases.

The results shown in Figure 3.3 are somehow like the trend on buckled arsenene. The only difference between the two is it has much lower thermal conductivity. From the range 10 nm up to 40nm, the results show that it has slowly increased, passes 40nm the thermal conductivity increase two times. The overall trend for both buckled and puckered arsenene conveys that, the thermal conductivity increases as the length increases.

As observed, when the length increased with an interval of 10nm, there are more present numbers of longer wavelength acoustic phonons which are free to participate in heat transfer in a phonon scattering. This employs that the thermal conductivity depends on the distance that phonon can travel between scattering events in their mean free path [20, 21].

This shows that the length dependence of thermal conductivity is controlled by phonon boundary scattering, while the length of buckled and puckered arsenene is governed by the phonon mean free path [20, 22]. As a result, when the length increases the phonon mean free path also increases since there is longer room for it to pass which corresponds to the increase of thermal conductivity [20]. Thus, developing a nanometer-scale 2D material can be applied to achieve an ultralow thermal conductivity, especially in electronic devices. Also, its fundamental understanding in terms of requirements in electronic devices is to explore the heat transport of material.

3.2. Temperature dependence of thermal conductivity

This section discusses the effect of temperature on the thermal conductivity of buckled and puckered arsenene. The result of thermal conductivity with varying temperatures in the range of 100K, 300K, 500, and 700K in a fixed dimension of 50nmx10nm was shown in Figure 3.4. Notice that, at the temperature of 100K up to 700K the thermal conductivity decreases, which shows similar results to the study in ab-initio and first-principle method [23, 24] and in monolayer 2D materials [21, 25]. This result also agrees with the thermal conductivity of the puckered arsenene shown in Figure 3.5

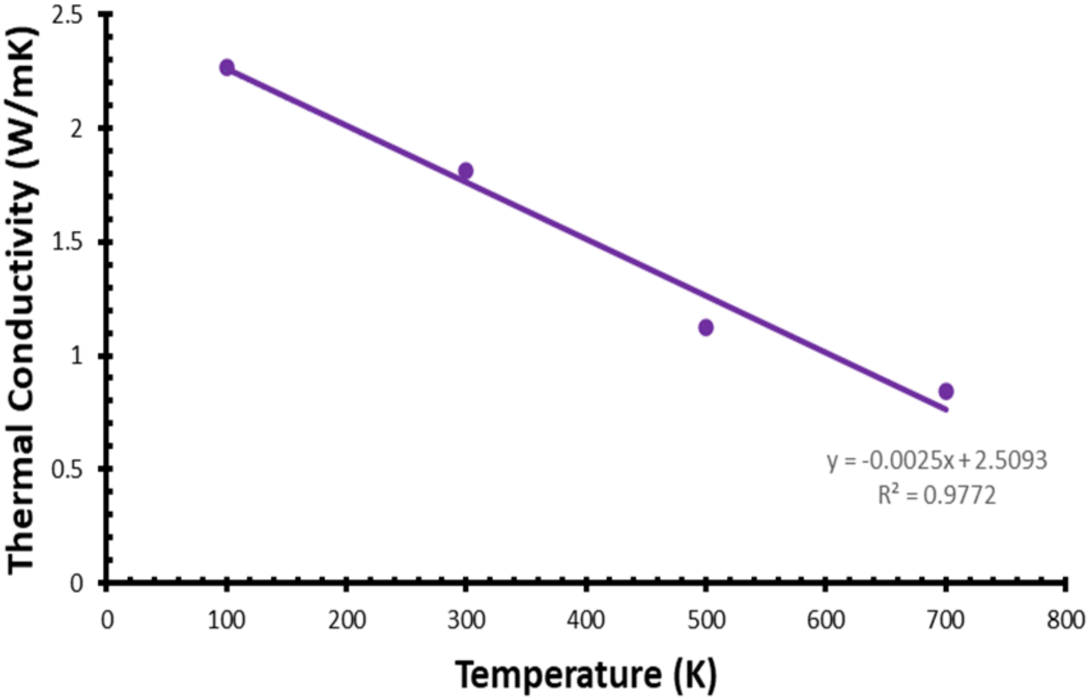

Figure 3.4. Thermal conductivity of Buckled arsenene as a function of temperature at a constant length of 50nm and width of 10nm. The trend shows decreasing thermal conduction when the temperature rises.

For this reason, lattice thermal conductivity has an inverse relationship with temperature [23]. At low temperatures, the thermal conductivity increases because of the phonons. The phonon mean free path almost become constant. This follows the specific heat, in which the thermal conductivity is proportional to the heat capacity. At high temperatures, the thermal conductivity decreases because heat capacity becomes constant which is equivalent to 3R where R is the universal gas constant. This satisfies the Dulong and Petitt law and expects that phonons undergo Umklapp scattering.

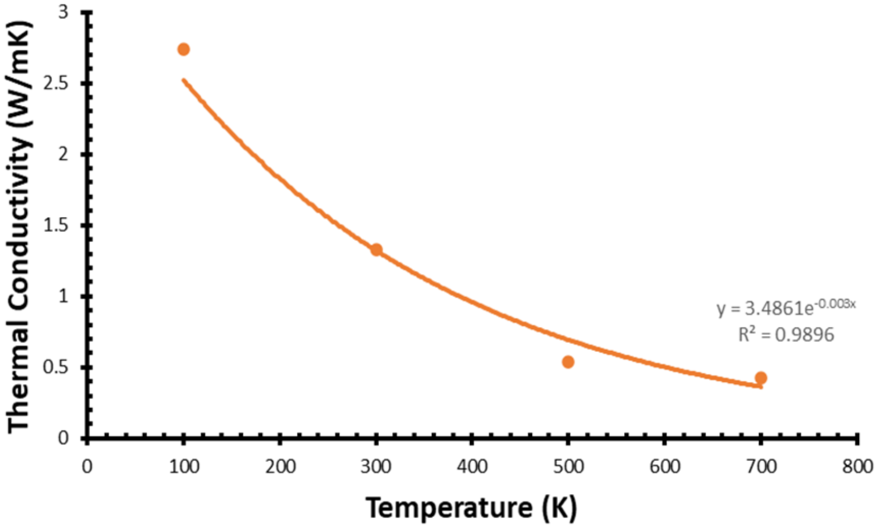

Figure 3.5. Thermal conductivity of Puckered arsenene as a function of temperature at a constant length of 50nm and width of 10nm. The trend shows decreasing thermal conduction when the temperature rises.

The phonon mean free path becomes smaller as the temperature rises which results in a decrease in thermal conductivity. Umklapp process is said to be inactive at low temperatures which cannot give a resistance that increases thermal conductivity. This is said to be the case for the decrease in thermal conductivity due to the growing resistance of Umklapp scattering [22]. The results show that the thermal conductivity for buckled arsenene at room temperature is found to have 1.8 W/mK and 1.3 W/mK for puckered arsenene.

4. CONCLUSION

It is found that when the length and temperature vary there is an increase in thermal conductivity on buckled and puckered arsenene. As the length increases, the presence of phonons that can participate is long enough to the increase phonon mean free path resulting in increased thermal conductivity. Moreover, the role of temperature in the system is found to be inversely proportional to thermal conductivity. The increase in temperature affects the phonon mean free path which becomes smaller and the decrease in thermal conductivity. The overall results of the thermal conductivity of buckled and puckered arsenene are found to be 1.8 W/mK and 1.3 W/mK, respectively at room temperature. As a result, the thermal conductivity of both buckled and puckered arsenene with varying length at fixed temperature increases highly in length 10nm-50nm. This is possibly due to the increased probability of collision as the dimension increases. It is speculated that the decrease in thermal conductivity as temperature rises is due to the Umklapp scattering process, where the phonon mean free path is inversely proportional to the temperature. The magnitude of phonon vibrations increases with temperature, increasing the probability of phonon-phonon scattering events. Anharmonic interactions between atoms become more significant at higher temperatures, resulting in phonon-phonon scattering processes.

At high temperatures, Umklapp scattering occurs, in which phonons change their wavevectors by interacting with lattice vibrations. Furthermore, lattice vibrations become more prominent, leading to greater lattice disorder, which impairs phonon coherent propagation, resulting in higher scattering and decreased thermal conductivity.

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