Study of Defects and their Impact on Transport and Thermoelectric Properties in Monolayer Silicene: an Ab Initio Simulation

Monalisa Ghosh*, Subhechcha Banerjee, Arnab Mukhopadhyay, Sahid Iqbal Mallick, Lopamudra Banerjee

Indian Institute of Engineering Science and Technology, Shibpur, India

*monalisaghosh12@gmail.com

Abstract: Here we present atomistic simulation study on the impact of defects on electronic transport and thermal properties of monolayer silicene. We have introduced four different type of defects in monolayer silicene sheet and these are single vacancy defects, stone-wales defect, edge roughness and ring missing defect. We have used Density functional theory (DFT) - Non equilibrium Green's function (NEGF) approach to study transport in pristine as well as in defective sheets. We have simulated the electron transmission and phonon transmission of the perfect and various defective sheets. We have simulated energy resolved transmission and I-V characteristics of pristine and defective sheets. Our simulation shows significant decrease in carrier conductance in the presence of defects as compared to pristine silicene sheet. Our investigation also shows a gradual decrease in current as the number of vacancy increases. We have computed electron conductance, peltier coefficient, Seebeck coefficient, heat transport coefficient for electron and phonon, thermal conductance and thermoelectric figure of merit ZT. Our computation presents a significant increase in ZT in presence of defects. ZT values for edge roughness, single vacancy defect, stone-wales defect are almost two times greater than pristine silicene sheet. Our simulation also shows a notable decrease in thermal conductance of silicene sheets for vacancy defects and stone-wales defect, edge roughness as compared to that for the perfect sheet. We have compared ZT of various defective sheets with pristine silicene sheet and this shows that edge rough silicene is better thermoelectric material as compared to perfect silicene.

Keywords: Silicene; defects; thermoelectric property; transmission spectra; density functional theory (dft).

1 INTRODUCTION

Graphene is a remarkable two dimensional material and discovery of its astonishing property has boosted further research in the area of two dimensional materials [1]. Intensified by the discovery of graphene various properties of nano-material analogous to graphene have been studied in a large scale which leads to exfoliation of

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material which can stand with improving technological requirement. In the wide variety of two dimensional materials, a special attention is given to monolayer silicon.

'Silicene' is a hexagonal structure of silicon atoms. As silicon is the most extensively used material in electronic applications, extensive research is appreciated in silicene to accelerate integration with electronic industry [2].

In the application field of monolayer material, defects are concerned topic. Various defects in two dimensional materials are introduced during fabrication process [3]. Vacancy defects, stone-wales defects, edge roughness etc are most common type of defects [4].

Some defects are almost unavoidable during fabrication process of monolayer material but sometimes defects are introduced deliberately for some specific application such as thermoelectric application, optoelectronic application etc. Recently, theoretical and experimental work is directed towards the impact of defects on electronic and thermal properties of monolayer silicene [5, 6].

Various researches on silicene present that rough silicon nanowire can be good thermoelectric material [7]. The measure for the performance of thermoelectric property of monolayer material is standardized by figure of merit ZT. $ZT=G_eS^2\,T/k$ where G_e is electrical conductance, S is Seebeck coefficient and T is temperature. The heat conduction k has both electronic and phononic component combined together, i.e $k= k_e + k_{ph}$ [8].

Although various studies have been conducted on thermoelectric properties of silicene but not much focus has been given to defects and their impacts on thermoelectric properties [8, 9].In this work, we present a computational study on the influence of various defects on thermoelectric properties of monolayer silicene. We consider four different types of defects such as single vacancy, ring missing, stone-wales defect and edge roughness. We have computed density functional theory in Quantum Wise ATK 2015.1 to investigate the impact of defects on electronic transmission and thermoelectric figure of merit. We have computed electron transmission spectra and evaluated current for pristine and defective sheets of monolayer silicene using DFT-NEGF method. We have also calculated phonon transmission spectra and evaluated ZT, Seebeck coefficient, conductance, peltier coefficient, thermal conductance for pristine and defective silicene sheets.

2 METHODOLOGY

In this work, we consider a super cell of monolayer silicene. The length of monolayer silicene sheet is 2.1 nanometer and width is 1.2 nanometer approximately. Fig. 1 shows various defects considered for our study.

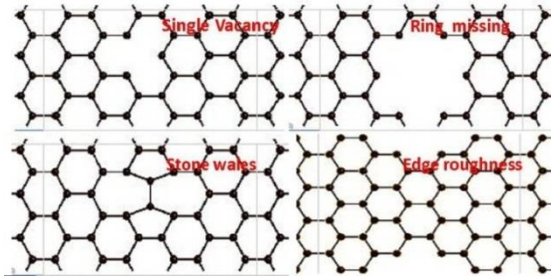

Fig. 1. Defects introduced.

2.1 Device Simulation

To calculate electronic transport and thermoelectric properties of the silicene super-cell under study, we apply density functional theory (DFT) in Quantum Wise ATK. Our calculation is performed using 3X3X1 Monkhorst-Pack-k-grid. We use Generalized Gradient Approximation (GGA)with Perdew Burke Ernzerhof Exchange (PBE) exchange correlation function with double Zeta polarized basis set and FHI(Z=4)GGA.PBE pseudo-potential set. We employ recursion as a self energy calculator and iteration control parameter algorithm is pulay Mixer (100 maximum steps and tolerance value of 0.0001 Hartree) [10]. Electron temperature is kept at 300K and a small bias 0.3 V is applied at right electrode.

In this work we have computed electron transmission coefficient [11]. Transmission formalism is also referred as Landauer approach where they assumed the channel to be connected between to contacts by two uniform leads. Landauer first developed the use of transport scattering theory. By this approach current can be calculated as [10, 11]

$$I=(q/h)\int_{-\infty}^{+\infty} dE\ T(E)[(f_0(E-\mu_1)-f_0(E-\mu_2)] \quad (1)$$

Current is zero at equilibrium as $\mu_1 = \mu_2$. An applied small bias voltage changes T(E), μ_1, μ_2 and current equation get modified as[10, 11]

$$I= (q/h)\int_{-\infty}^{+\infty} dE\ \ \delta T(E)[(f_0(E-\mu_1)-f_0(E-\mu_2)] + (q/h)\int_{-\infty}^{+\infty} dE\ T(E)\delta[(f_0(E-\mu_1)-f_0(E-\mu_2)] \quad (2)$$

The first term in equation 2 is zero and the second can be written as [10, 11]

$$I \approx \left(\frac{qV^2}{h}\right) \int_{-\infty}^{+\infty} dE\, T(E)(-df_0(E)/dE)_{E=\mu} \quad (3)$$

Conductance G is

$$G = (q^2/h)T_0 \quad (4)$$

Where

$$T_0 = \int_{-\infty}^{+\infty} dE\, T(E)F_T(E-\mu) \quad (5)$$

Current is independent of voltage application i.e whether it is applied in right or left electrode, it only depends on their difference i.e V_{bias}. Here $V_{bias} = 0.3\ V$.

We have also calculated the phonon transmission coefficient to investigate phonon thermal conductance which is required to obtain ZT. Phonon transmission coefficient is combined with electron transmission to obtain conductance, peltier coefficient, and electron thermal conductance. These parameters are combined to obtain thermoelectric figure of merit ZT. We have computed linear transport coefficient, ZT at an applied voltage difference dU between two electrodes in the silicene device [12, 13].

Conductance G_e is calculated as [14]

$$G_e = \frac{dI}{dU}\Big|_{dT=0} \quad (6)$$

Where dT is the temperature difference.

The peltier coefficient is calculated as [14]

$$\Pi = \frac{I_Q}{I}\Big|_{dT=0} \quad (7)$$

The Seebeck coefficient is calculated as [14].

$$S = -\frac{dU}{dT}\Big|_{I=0} \quad (8)$$

The heat transport coefficient of electron k_e and phonon k_{ph} [14]

$$K = \frac{dI_Q}{dT}\Big|_{I=0} \quad (9)$$

Where $I_Q = dQ/dT$ is the heat current. T is the temperature.

From these above equations thermoelectric figure of merit (ZT) can be calculated [14].

$$ZT = G_e S^2\, T/k \quad (10)$$

From equation (10) it is shown that ZT can be increased by increasing the power factor $G_e S^2\, T$ or decreasing the thermal conductance k. Thus to exhibit high thermoelectric performance, a material should posses a high value of seebeck coefficient, electrical conductance as well as low thermal conductance.

3 RESULTS AND DISCUSSIONS

Figure 2 shows Transmission spectra for pristine silicene and various defective sheets of silicene. We observe that the transmission spectra computed in the range of -3 to 3 eV decreases in the presence of defects. Transmission spectra for ring missing defect is almost negligible with respect to transmission spectra of perfect silicene sheet.

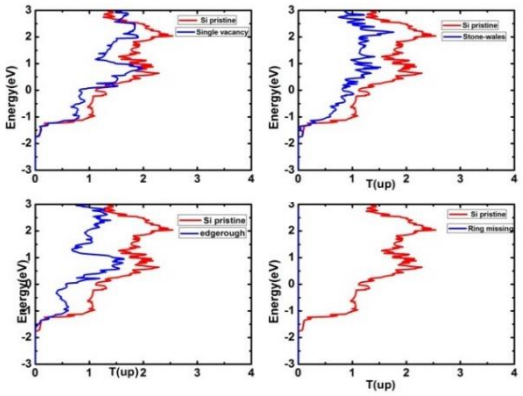

Fig. 2. Transmission spectra of defective silicene sheets in comparison to pristine silicene sheet.

Transmission peaks for pristine and defective silicene sheet are obtained at different energy values which are shown in Table 1.

Table 1. Energy values where maximum transmission coefficient are obtained for silicene sheets.

Sheet	Energy (eV)	Transmission
Pristine	2.04	2.5441
Single Vacancy	0.88	1.94786
Edge Roughness	0.98	1.57588
Ring missing	1.08	6.562e-6
Stone-wales	2.18	1.7638

Figure 3 represents the current obtained for pristine as well as defective sheets of monolayer silicene.

It can be inferred from Fig. 3 that pristine silicene carries maximum current and current for defective sheets are smaller than the perfect sheet and as number of vacancy increases in the sheet current gradually decreases.

Fig. 3. Variations of Current with defects in silicene sheet.

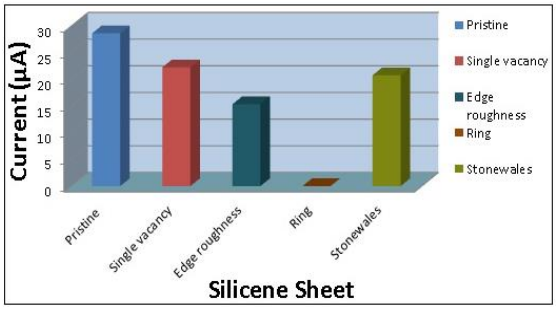

Fig. 4 shows thermoelectric figure of merit ZT for perfect and defective silicene sheets.

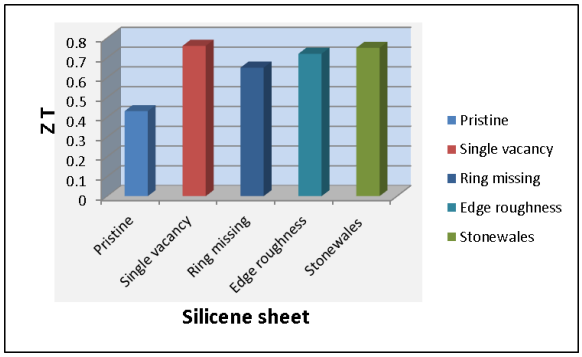

Fig. 4. Variations of ZT with defects in silicene sheet.

We can see that ZT is least for pristine silicene sheet and ZT increases with the introduction of defects. We have studied that material with ZT~1 is considered as good thermoelectric. In presence of defects in silicene sheet ZT is tending to 1. Hence, It can be inferred that higher ZT can be obtained in case of defective sheets as compared to pristine silicene sheet. This is due to opening up of more conduction channels near the fermi level due to defect states.

Fig. 5 presents the thermal conductance for electron (k_e), phonon (k_p) and total thermal conductance (k) of pristine silicenesheet.

Figs. 6-9 shows the thermal conductance of defective silicene sheets. From the figures of thermal conductance for pristine and defective we see that thermal conductance peak is maximum for pristine silicene and it decreases in presence of defects. Thermal conductance is minimum for ring missing defect. Thermal conductance gradually decreases as the number of vacancy increases.

Table 2 shows electronic thermal conductance, phononic thermal conductance and total thermal conductance at zero eV energy. The total thermal conductance is the summation of electronic and phononic thermal conductance. We can observe that at zero energy value total thermal conductance is maximum for pristine silicene sheet.

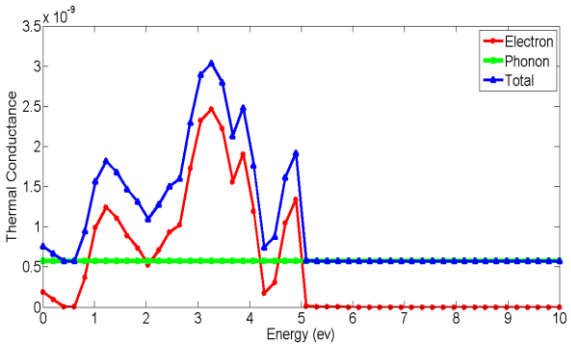

Figure 5. Thermal conductance of pristine silicene.

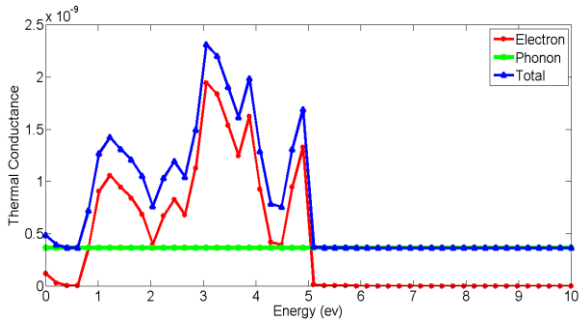

Figure 6. Thermo electric conductance of single vacancy defect.

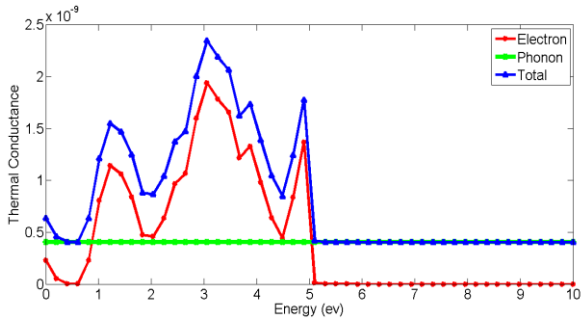

Figure 7. Thermal conductance for edge roughness.

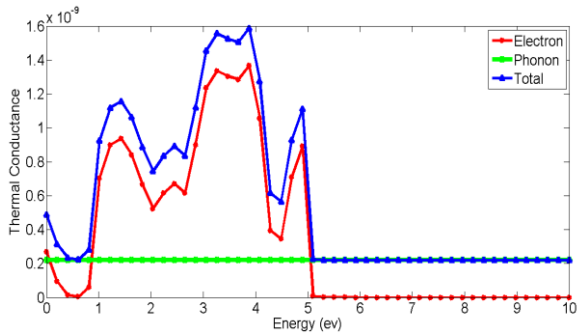

Figure 8. Thermal conductance for ring missing defect.

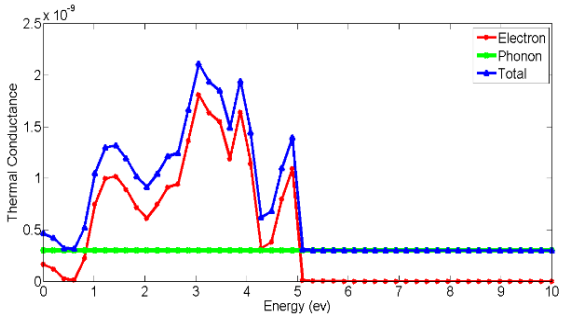

Fig. 9. Thermal conductance for stonewales defect.

Table 2. Thermoelectric conductance for electron, phonon and the total for pristine and various defective silicene sheets at Energy=0 eV.

Sheet	Electron (k_e)	Phonon (k_{ph})	Total (k)
Pristine	1.875Xe^{-10}	5.712Xe^{-10}	7.587Xe^{-10}
Single Vacancy	1.203Xe^{-10}	3.646Xe^{-10}	4.849Xe^{-10}
Stone-wales	1.629Xe^{-10}	3.002Xe^{-10}	4.631Xe^{-10}
Edge Roughness	2.29Xe^{-10}	4.061Xe^{-10}	6.351Xe^{-10}
Ring missing	2.672Xe^{-10}	2.199Xe^{-10}	4.871Xe^{-10}

Table 3 lists various thermoelectric coefficients such as conductance, peltier coefficient, Seebeck coefficient for perfect and four different defective silicene sheets.

Table 3. Thermoelectric parameters (conductance values, peltier coefficient and Seebeck coefficient) obtained for Silicene sheets.

Sheet	Conductance (G_e)	Peltiercoeff (Π)	Seebeck coeff (s)
Perfect	3.87x10-5S	0.011581	5.27x10-5
Single vacancy	4.21x10-5S	0.002054	-6.84x10-6
Ring missing	3.83x10-5S	-0.006618	-2.20x10-10
Stone-wales	2.17x10-9S	0.01432	4.77x10-5
Edge roughness	4.17x10-5S	0.01043	3.47x10-5

4 CONCLUSION

Here, we have investigated the impact of defects on transport and thermoelectric properties of monolayer silicene sheets. Various defects like single vacancy, ring missing defect, stone-wales defect and edge roughness are introduced and studied with DFT.

We observe that the transmission peak decreases in the presence of defects .Carrier conductance reduces significantly in the presence of defects as compared to the pristine sheet. Our investigation shows that thermoelectric figure of merit ZT increases with introduction of defect in monolayer silicene sheet (suggesting defects, edge roughness make silicene a better thermoelectric material as compared to pristine silicene sheet). Thermal conductance degrades significantly in the presence of defects .Thermal conductance is maximum for pristine silicene sheet and minimum for ring missing defect.

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