Impressive Electronic Transport in Be$_2$C
Monolayer Limited by Phonon

Gautam Sharma*,,† and K.C. Bhamu‡

†Department of Physics, Indian Institute of Science Education and Research Pune,
Pune-411008, India
‡PMC Division, CSIR-National Chemical Laboratory, Pune-411008, India

E-mail: gautam.iiser@gmail.com,kcbhamu85@gmail.com

Abstract

We present thermoelectric properties of Be$_2$C monolayer based on density functional theory based calculations combined with semi-classical Boltzmann transport theory. First principles calculations show the material is direct band gap semiconductor with band gap of 2.0 eV obtained with Gaussian-attenuating Perdew-Burke-Ernzerhof (Gau-PBE) hybrid functionals. Kohn-Sham eigen-states obtained with Gau-PBE are fed into Boltzmann transport equation which is solved under constant relaxation time approximations resulting into thermoelectric (TE) coefficient in terms of constant relaxation times ($\tau$). In this work, we have explicitly determined the relaxation time by studying the electron-phonon interactions by means of electron-phonon coupling using Wannier functions package to obtain the absolute TE coefficients along armchair and zigzag directions. Our results shows that material has high TE coefficients like Seebeck coefficient ($\alpha$) and electrical conductivity ($\sigma$) leading to high power factor ($\alpha^2\sigma \sim 3.44$ mW/mK$^2$) along zigzag direction with hole doping which is found in the similar order of the PF reported for commercial TE materials.(J. Appl. Phys. 2003 (93) 368-374; J. Appl. Phys. 2008 (104) 053713-1-053713-5). Further, third-order anharmonic theory
reveals the slightly high lattice thermal conductivity (∼ 66 W/mK) at 300 K giving rise to moderate values of ZT (∼ 0.1) optimized with p-type doping along zigzag direction. Our results suggest that Be₂C monolayer is promising material for thermoelectric applications as far as high power factor is concerned. Additionally, the dynamical stability of the Be₂C monolayer up to 14% bi-axial strain shows that phonon transport in the Be₂C monolayer can be further improved through strain engineering.

1 INTRODUCTION

Increasing consumption of the natural energy resources demanded the scientific community to switch their attention to sustainable energy resources. Waste heat management technology is one of the efficient and economic alternative for power generation. Thermoelectric (TE) materials have gained a huge amount of attention for ability to convert the waste heat ejected from automobiles and industries and convert it into electricity. Figure of merit, ZT = α²σT/κ, is the key quantity which describe the efficiency of a TE material, where T, α, σ and κ are temperature, Seebeck coefficient, electrical conductivity and total thermal conductivity, respectively. Thermal conductivity arises from combination of the electronic and lattice thermal conductivity. (κ = κₑ + κₙ). ZT can be maximized by increasing the power factor (PF, PF=α²σ) and simultaneously decreasing the thermal conductivity (κ). However, achieving high PF by increasing σ results in increase of κₑ since they are connected by Wiedemann-Franz Law. In contrast κₙ is independent of σ, thus performance can be enhanced by decreasing κₙ which contributes significantly to κ. TE performance of the material can be tuned by various ways like convergence of multiple bands, combination of light-heavy bands, multivalley carrier pocket and reduced dimensionality. Hicks et al. have shown theoretically that low-dimensional quantum systems would attain higher ZT than their bulk counterpart because of reduction in the characteristic length of phonon by adjusting the sample size. Recent advances to improve ZT are centered around reducing dimensionality, for instance, monolayer Bi₂Te₃ have shown high ZT than its bulk analogs.
Similarly, nanostructured materials have also shown better TE performance.\textsuperscript{12}

In this work, we study thermoelectric properties of Be\textsubscript{2}C monolayer (Be\textsubscript{2}C-ML) in its Global minimum energy structure using first principles density functional theory (DFT) based calculations combined with Boltzmann transport theory (BTT). Be\textsubscript{2}C-ML is found to be a direct band gap material of 1.52 and 2.34 eV with Perdew, Burke and Ernzerhof (PBE) and Heyd-Scuseria-Ernzerhof (HSE) approaches, respectively.\textsuperscript{13} In a computational study, Naseri \textit{et al.} have investigated electronic and optical properties under stress and strain.\textsuperscript{14} Yeoh \textit{et al.} have studied Be\textsubscript{2}C-ML as the anode material for Lithium-ion battery applications.\textsuperscript{15} Electronic band structure shows that it has doubly degenerate valence bands at the $\Gamma$ point. These bands provide both light and heavy charge carriers to enhance the transport properties.\textsuperscript{13} Moreover, studies using molecular dynamics (MD) has shown that it has a high melting point around 1500 K and therefore, it has a potential to perform even at high temperatures.\textsuperscript{13} To best of our knowledge, TE properties of Be\textsubscript{2}C are still waiting to uncover. These factors have motivated us to investigate the TE properties of Be\textsubscript{2}C-ML. Furthermore, the electron-phonon (el-ph), phonon-phonon (ph-ph) interactions within Be\textsubscript{2}C-ML and lattice thermal conductivity of the material are not yet studied. So we also investigate the aforementioned scattering mechanisms occurring in the system. The rest of manuscript is organized as follows. Section 2 provides the details of computational methods used to investigate various properties. Section 3 discusses electronic structure, el-ph scattering, TE properties, ph-ph scattering and lattice thermal conductivity of the Be\textsubscript{2}C-ML. Conclusive remarks are presented in Section 4.

2 COMPUTATIONAL DETAILS

\textbf{First principles calculations:} First principles calculations are performed with the Quantum ESPRESSO software, which is a plane wave based implementation of DFT.\textsuperscript{16,17} The electronic exchange and correlation potential is described by the generalized gradient ap-
proximation (GGA) as parametrized by Perdew, Burke and Ernzerhof (PBE).\textsuperscript{18} We have used the norm-conserving Trouiller-Martins pseudopotentials to describe the electron-ion interactions for all the calculations.\textsuperscript{19} The pseudopotentials have been generated using 2s\textsuperscript{2} 2p\textsuperscript{2} 3d\textsuperscript{0} 4f\textsuperscript{0} and 2s\textsuperscript{2} 2p\textsuperscript{2} 3d\textsuperscript{0} 4f\textsuperscript{0} valence configurations for Be and C respectively. The wave functions are expanded in a plane wave basis, the size of which is defined by the kinetic energy cut-off of 85 Ry. To sample the Brillouin zone (BZ), we have chosen a Monkhorst-Pack (11×11×1) k-mesh for the monolayer.\textsuperscript{20} We have included semi-empirical van der Waals (vdW) corrections in all the calculations as proposed by Grimme.\textsuperscript{21} For better estimate of band dispersion, we have performed hybrid calculations using the singularity free Gaussian-attenuating Perdew-Burke-Ernzerhof (Gau-PBE) hybrid functionals.\textsuperscript{22} The matrix elements of the Fock operator is evaluated with a (5×5×1) q-mesh for the ML. We have used a vacuum of length L, 15 Å, to eliminate the periodic image interactions present along z-axis.

**Relaxation time (τ) calculations:** Within Migdal approximation, the imaginary parts of electron self-energy ($\Sigma_{n,k}'=\Sigma_{n,k}'+i\Sigma_{n,k}''$) is given by.\textsuperscript{23,24}

\begin{equation}
\Sigma_{n,k}'' = \sum_{\nu q} w_q |g_{\nu mn}|^2 \left[ \frac{n(\omega_{\nu q}) + f(\epsilon_{mk+q})}{\epsilon_{nk} - \epsilon_{mk+q} - \omega_{\nu q} - i\eta} + \frac{n(\omega_{\nu q}) + 1 + f(\epsilon_{mk+q})}{\epsilon_{nk} - \epsilon_{mk+q} - \omega_{\nu q} + i\eta} \right]
\end{equation}

where $w_q$ is weight associated with q in the BZ, $g_{\nu mn}'$ is el-ph matrix element. $n(\omega_{\nu q})$ and $f(\epsilon_{mk})$ are the Bose and Fermi occupation factors respectively. $\omega_{\nu q}$ is the phonon frequency for mode $\nu$ and wave vector q and $\epsilon_{nk}$ are the Kohn-Sham eigen values for band n and wave vector k. The imaginary part of self-energy of the electron (Im$\Sigma_{n,k}$) is computed with the EPW (Electron-Phonon coupling using Wannier functions) package.\textsuperscript{25} The relaxation time ($\tau_{n,k}$) is inversely related to Im$\Sigma_{n,k}$ as follows:

\begin{equation}
\tau_{n,k} = \frac{\hbar}{2\text{Im}\Sigma_{n,k}}
\end{equation}

where $\hbar$ is the reduced Planck constant. We calculate electron self-energy using PBE functionals, firstly, we compute the electronic structure on a coarse (18×18×1) k-grid which
is further interpolated using maximally localized Wannier functions (MLWFs).\textsuperscript{26} Then we compute phonon dispersion and el-ph matrix elements ($g_{mn}^\nu$) on a coarse (9×9×1) q-grid by density functional perturbation theory (DFPT).\textsuperscript{27} To obtain converged results, the coarse k and q-grids are interpolated on a (25×25×1) k-grid and a dense (280×280×1) q-grid respectively.

**Boltzmann Transport calculations:** Thermoelectric coefficients such as Seebeck coefficient ($\alpha$), electrical conductivity ($\sigma$) can be calculated using semi-classical Boltzmann Transport equation (BTE).\textsuperscript{28} BoltzTraP package is used to solve the BTE to provide TE coefficients and it considers the rigid band approximation (RBA) and constant relaxation time approximations (CRTA).\textsuperscript{29} Within RBA, effects on the band energies are ignored if the system is either doped or temperature of the material changes and the chemical potential ($\mu$) is shifted above/below for electron/hole doping. Here, we use a dense (35×35×1) k-grid for electronic structure calculations performed with Gau-PBE functionals to achieve convergence in transport coefficients.

**Calculation of lattice thermal conductivity ($\kappa_l$):** The temperature-dependent lattice thermal conductivity ($\kappa_l$) is calculated by solving BTE for phonons as implemented in ShengBTE code\textsuperscript{30} using second and third-order interatomic force constants (IFCs). We obtain second order IFCs with (5×5×1) q-mesh by DFPT as implemented in Quantum ESPRESSO code\textsuperscript{27} and third-order anharmonic IFCs are generated using thirdorder.py script provided in ShengBTE using (5×5×1) supercell size and (3×3×1) k-mesh. The effect of nearest-neighbors interaction are seen on anharmonic IFC starting from third-nearest neighbors to fifth-nearest neighbors and results converge by considering the third-nearest neighbors interactions. We have used a dense (90×90×1) q-mesh to ensure the convergence of $\kappa_l$. Further we have scaled $\kappa_l$ by ratio of vacuum length L to the effective thickness ($d_{eff} = 4.19$ Å) of the monolayer such that the volume of the parallelepiped generated by in-plane lattice vectors and $d_{eff}$ accommodates 99 % of the total charge (Q). We calculate the total charge by integrating the planar average of charge density ($\bar{\rho}(z) = \frac{1}{A} \int_A \rho(x,y,z)dx dy$) using the
3 RESULTS AND DISCUSSIONS

3.1 Crystal structure of Be$_2$C-ML

Fig. 1(a) shows the hexagonal crystal structure of Be$_2$C-ML with space group (164) P-3m1. The Be$_2$C-ML comprises of three atomic sublayers, one C-layers and two Be-layers stacked along z-direction. The C layer is sandwiched between two Be layers on either side. Within Be$_2$C-ML, each C atom is connected with six Be atoms and each Be atom is connected with three C and three Be atoms. The optimized Be-C and Be-Be bond lengths are 1.76 Å and 1.93 Å respectively which agrees well with previous report. The intralayer distance between the two Be layers is $\Delta \sim 0.91$ Å and that between the C and the Be layer is $\sim 0.45$ Å as obtained from the present study. Our calculations with PBE functionals yield lattice parameter, $a = 2.95$ Å, and these are in excellent agreement with the previously reported value $2.99$ Å.

![Figure 1](image.png)

Figure 1: (a) Top (b) side view of unit cells for Be$_2$C-ML. Beryllium atoms are represented by dark blue (top) and light green (bottom) spheres and the carbon atoms present in the middle layers are represented by brown spheres.
3.2 Electronic band structure and density of states

The PBE band structure of Be$_2$C-ML (red solid lines) with vdW corrections is shown in Fig. 2(a). We observe, it has a direct band gap of 1.62 eV with the valence band maxima (VBM) and the conduction band minima (CBM) located at Γ point. Since PBE functional are known to underestimate the band gaps, therefore we calculate the band structure with Gau-PBE hybrid functionals with vdW corrections to predict the accurate band gap. These functionals provide Kohn-Sham states as accurate as can be obtained by HSE hybrid functionals with very less computational cost. We find that Gau-PBE predicts the band gap of 2.0 eV whereas the band dispersion remains intact compared to PBE as shown in Fig. 2(a) (green solid lines). The computed band gap is 0.34 eV lower than the previous report obtained with HSE. The total density of states (DOS) with PBE and those projected onto the Be-$s,p,d$ and C-$p,d$ atomic orbitals are shown in Fig. 2(b). We see the valence band is primarily composed of the C-$p$ and Be-$s,d$.

![Band structure and DOS](image)

Figure 2: (a) Band structure for Be$_2$C-ML with PBE (red) and Gau-PBE (green) functionals. (b) Projected density of states (PDOS) with PBE (b). The vacuum energy is chosen as zero to shift the Kohn-Sham energy states.

3.3 Electron-Phonon Relaxation time ($\tau$)

The TE coefficients are generated in terms of relaxation time ($\tau$), namely, electrical ($\sigma/\tau$) and electronic thermal conductivity ($\kappa_e/\tau$) when they are computed within Boltzmann transport framework under CRTA. Therefore, it is of prime importance to calculate $\tau$ of the charge
carriers within the monolayer explicitly to evaluate the absolute TE coefficients. Hence we exploit the most advance methods, EPW, based on the el-ph interactions where acoustic and optical vibrational modes are taken to be scattering channels. As a result, it precisely provides information of τ compared to traditionally used deformed potential (DP) theory wherein scattering only from acoustic modes is considered and as a result, it severely over-estimates the TE coefficients.11,31,32

![Figure 3: (a) Variations of imaginary part of self-energy for valence ($\Sigma_{VB}$) and conduction bands ($\Sigma_{CB}$) (solid and dashed lines respectively) along the high symmetry directions in the BZ at 300K, 500K and 700K (red, blue and green respectively) for the Be$_2$C-ML. (b) Temperature-dependent average relaxation time ($\tau_{av}$) for electron (red) and holes (blue).](image)

Fig. 3 (a) shows variation of relaxation rate ($1/\Sigma_k$) for the top conduction (solid lines) and the valence bands (dashed lines) along the high symmetry directions in the BZ for the temperatures: 300K, 500K and 700K at Fermi energy. We find that it increases with increase in the temperature leading to decrease in $\tau_{av}$ with temperature as shown in Fig. 3 (b). Here $\tau_{av}$ is obtained for conduction ($\tau_e$) and valence bands ($\tau_h$) by summing $\Sigma_k$ over all k-states in the entire BZ at a given temperature using ($\tau_n = \sum_k \frac{\epsilon_{n,k}\tau_{n,k}}{\epsilon_{n,k}}$).33 Similarly, $\tau_{av}$ can also be obtained along zigzag and armchair directions by supplying path along the M-K and $\Gamma$-M directions in BZ, respectively. Having the information of the $\tau_{av}$ for holes/electrons at various temperatures, we compute the transport properties of the Be$_2$C-ML as explained in the following section.
3.4 Transport properties

Transport properties of Be$_2$C-ML are produced as a function of electron/hole doping at three different temperature values. Notice that we compute TE properties, namely, electrical conductivity ($\sigma$) and electronic thermal conductivity ($\kappa_e$) along zigzag and armchair directions by plugging in the $\tau_{av}$ obtained along respective directions as mentioned in the previous section. Since the Seebeck coefficient ($\alpha$) is obtained independent of $\tau$ within Boltzmann transport framework under CRTA. Therefore, it is expected to be same along the two directions.

**Transport along zigzag direction with p(n)-type doping:**

In Fig. 4 (a) we plot magnitude of Seebeck coefficient (|$\alpha$|) as a function of carrier concentration at different temperatures for holes (solid lines) and electron doping (dashed lines). We find that the $\alpha$ decreases as the carrier concentration increases and rises with ramp up of the temperature. The value of $\alpha$ (at optimum PF obtained with hole doping) is found to be 0.167 mV/K with hole doping of $\sim 6.6 \times 10^{12}$ cm$^{-2}$ at 300 K and it is of similar order as observed for Bi$_2$Te$_3$. Notice that Seebeck is greater for electron than holes due to differences in effective masses of the carriers involved. The value of $\alpha$ for the electron doping is -0.167 mV/K at the carrier concentration of $1.2 \times 10^{13}$ cm$^{-2}$ at 300 K where the PF tends to optimum value with electron doping. Fig. 4 (b) depicts the electrical conductivity ($\sigma$) for the monolayer with hole (electron) doping. The $\sigma$ decreases with increase in temperature since relaxation time decreases with temperature for both holes and electrons. We have obtained the value of $\sigma$ (at optimum PF) to be $1.22 \times 10^5$ S/m with hole doping of $6.6 \times 10^{12}$ cm$^{-2}$ at 300 K. Fig. 4 (c) presents electronic contribution to thermal conductivity ($\kappa_e$) with hole/electron doping. We find that $\kappa_e$ is directly proportional to electrical conductivity which is typical trends among them as suggested by Wiedemann-Frenz law. Fig. 4 (d) shows the PF for the monolayer when it is hole/electron doped. As the power factor is function of $\alpha$ and $\sigma$ where both shows opposite trends when monolayer is doped. Therefore, one is expected to find the optimum value of PF as function of carrier concentration. For hole
doped of $6.6 \times 10^{12}$ cm$^{-2}$, we have obtained the optimum value of PF to be $\sim 3.44$ mW/mK$^2$ at 300 K which is of same order observed for bulk Bi$_2$Te$_3$.$^{34,35}$ Further we see that the PF increases as temperature increases due to increase in $\alpha$. The value of optimum PF is $0.92$ mW/mK$^2$ at the electron doping of $1.2 \times 10^{13}$ cm$^{-2}$ and it is almost four times lower compared to PF obtained for hole doping due lower electrical conductivity of the electrons.

![Zigzag](image)

Figure 4: The Seebeck coefficient (a) electrical conductivity (b) power factor (c) electronic thermal conductivity (d) with hole (electron) doping along zigzag direction is shown by solid (dashed) lines at 300 K, 400 K and 500K represented by black, red and blue lines respectively.

Transport along armchair direction with p(n)-type doping: Fig. 5 (a-d) depicts the TE coefficients along armchair direction for hole/electron doping. TE properties for armchair show similar trends with hole/electron doping as observed along zigzag direction. However, transport properties have smaller magnitudes along the armchair direction compared to zigzag direction. The difference in the magnitude of TE coefficients arise due to differences in the lifetimes of the charge carrier in the two directions.
Figure 5: The Seebeck coefficient (a) electrical conductivity (b) power factor (c) electronic thermal conductivity (d) with hole (electron) doping along armchair direction is shown by solid (dashed) lines at 300 K, 400 K and 500K represented by black, red and blue lines respectively.

3.5 Lattice thermal conductivity ($\kappa_l$) and figure of Merit:

Fig. 6 (a) illustrates the phonon spectrum of Be$_2$C-ML computed for unstrained system (green). The phonon dispersion for unstrained Be$_2$C-ML agrees well the previous reports.13,27 Be$_2$C-ML has a total of 9 modes of vibration: first three, lying in lower frequency region, are acoustic modes and others are called optical modes. According to vibrational patterns, modes are named as flexural acoustic (ZA), Transverse acoustic (TA) and Longitudinal acoustic modes (LA), where ZA modes describes atoms vibrating out-of-plane direction and the other two modes describing the motion of atoms in-plane of monolayer at equilibrium but parallel (LA) and perpendicular (TA) to speed of sound. In the vicinity of zone center, ZA branch is proportional to $q^2$ for the unstrained system and other acoustic branches are proportional to $q$. For unstrained system, we have obtained the sound velocities to be 1616 ms$^{-1}$ and 1133 ms$^{-1}$ from the slopes of LA and TA branch near $\Gamma$, respectively. These values are much lower than the sound velocities found in the graphene$^{36}$ and black phospho-
Therefore, we expect to find much lower values of $\kappa_l$ for the Be$_2$C-ML as compared to graphene and black phosphorene.

Further, we obtain the lattice thermal conductivity for the unstrained system by solving the linearized BTE for phonons using iterative methods at different temperature values. In this work, we have considered two scattering mechanisms, namely, anharmonic three ph-ph and isotopic scattering. The phonon group velocities as a function of $\omega$ are depicted in Fig. 6 (b) for all the modes. We find that lattice thermal conductivity of Be$_2$C-ML is isotropic and computed to be $\sim 66$ W/mK at 300 K. The value of $\kappa_l$ is already lower than observed for other 2D materials like MoS$_2$ and Graphene$^{38,39}$

Phonon engineering is the high efficient approach to reduce the $\kappa_l$ and thus to improve the ZT.$^{40}$ To see scope of reducing the $\kappa_l$ in Be$_2$C-ML, we have introduced strain in the monolayer and found that up to 14 % of applied bi-axial strain it remains dynamically stable, Fig. 6 (a) (red). The applied strain significantly reduced the velocities of LA and TA modes.
to be 800 ms$^{-1}$ and 517 ms$^{-1}$, respectively. It is well known that $\kappa_l$ is proportional to square of phonon group velocities.$^{30}$ Thus lower values of group velocities for Be$_2$C-ML will further significantly reduce the $\kappa_l$ and similar has been observed in ScP and ScAs monolayers.$^{41}$ Moreover, strain also flattens the optical phonon bands relative to the unstrained case and thus the contribution of optical bands to $\kappa_l$ is further reduced in strained case. Further, we find $\kappa_l$ decreases with increasing temperature as depicted in Fig. 6 (d). This is because three-phonon scattering rate increase with rise in temperature as shown in Fig. 6 (c). The fitting of $\kappa_l$ with $T^{-1}$

![Graph](image)

Figure 7: The variation in ZT as a function to hole (electron) doping concentration at 300-800K with blue (red) lines along zigzag direction.

Fig. 7 depicts optimum values of thermoelectric figure of Merit (ZT) for Be$_2$C-ML along zigzag direction with the hole (blue) and electron (red) doping as function of temperature. We notice that the values of ZT are larger with p-type doping than n-type. We obtain the ZT which is around 0.14 (0.04) at 800 K with hole (electron) doping of $1.6(3.0) \times 10^{13}$ cm$^{-2}$ and it is of similar order observed for 2D materials like MoS$_2$ and phosphorene suggesting the novelty of Be$_2$C-ML in TE applications.$^{31}$
4 CONCLUSION

In conclusion, we have computed electronic band structure and TE properties with Gau-PBE hybrid functionals. We find Be$_2$C-ML is direct band gap material with doubly degenerate energy states at $\Gamma$ as desirable for TE applications. We have used most advanced method, EPW, to calculate the average relaxation times for charge carriers which is then incorporated in TE coefficients to predict most accurate PF. The obtained value of PF ($\sim 3.44$ mW/mK$^2$ at 300 K) is found to be of the similar order of PF as reported for many well known TE materials like Bi$_2$Te$_3$ which reflects that Be$_2$C-ML may be a potential room temperature TE material. The calculated phonon lattice thermal conductivity ($\sim 66$ W/mK at 300 K) is found tolerable in comparison to graphene and other 2D materials. Notably, dynamical stability of Be$_2$C-ML up to 14 % bi-axial strain reduces the phonon group velocities by $\sim 50\%$ compared to unstrained case which could further reduce $\kappa_l$ by 75% at 300 K which in turn make the Be$_2$C-ML an ultra low $\kappa_l$ material meeting the low $\kappa_l$ criteria of commercial TE material. Finally, for unstrained system we have obtained the $ZT_{max} \sim 0.1$ with hole doping $1.6 \times 10^{14}$ cm$^{-2}$ at 800 K along zigzag direction and which is limited by phonon transport but can certainly be tuned via strain engineering.

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