Carbosilicene and germasilicene: Two 2D materials with excellent structural, electronic and optical properties

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Using first principle calculations, we study the structural, optical and electronic properties of two-dimensional silicene-like structures of CSi7 (carbosilicene) and GeSi7 (germasilicene) monolayers. We show that both CSi7 and GeSi7 monolayers have different buckling that promises a new way to control the buckling in silicene-like structures. Carbon impurity decreases the silicene buckling, whereas germanium impurity increases it. The CSi7 has semiconducting properties with 0.25 eV indirect band gap, but GeSi7 is a semimetal. Also, under uniaxial tensile strain, the semiconducting properties of CSi7 convert to metallic properties which shows that CSi7 can be used in straintronic devices such as strain sensor and strain switch. There is no important response for GeSi7 under strain. The GeSi7 has higher dielectric constant relative to CSi7, silicene and graphene and it can be used as a 2D-material in high performance capacitors. Calculation of cohesive and formation energies show that CSi7 is more stable than GeSi7. Furthermore, we investigate the optical properties of these new materials and we show that CSi7 and GeSi7 can significantly increase the light absorption of silicene. The obtained results can pave a new route for tuning the electronic and optical properties of silicene like structures for different applications in nanoelectronic devices.

I. INTRODUCTION

Two dimensional (2D) materials like graphene, silicene, and germanene are semimetals with zero-gap [1, 2], and their charge carriers are massless fermions [3]. Graphene have been studied vastly because of its superior advantages such as mechanical, optical and electronic properties [4–18]. Different doping are performed in graphene for the new applications such as sulfur-doping for micro-supercapacitors [19], nitrogen-doping are performed in graphene for the new applications about doped atoms such as lithium, aluminum and phosphorus to control the electronic and optical properties. Here we call CSi7 or a semiconductor. g-SiC3 has a good structural, electronic and optical properties. For example, SiC2 siligraphene which has been experimentally synthesized [24], is a promising anchoring material for lithium-sulfur batteries [37], a promising metal-free catalyst for oxygen reduction reaction [37], and a novel donor material in excitonic solar cells [38]. Also, graphitic siligraphene g-SiC3 in the presence of strain can be classified in different electrical phases such as a semimetal or a semiconductor. g-SiC3 has a semimetallic behavior under compression strain up to 8%, but it becomes a semiconductor with direct band gap (1.62 eV) for 9% of compression strain and becomes a semiconductor with indirect band gap (1.43 eV) for 10% of compression strain [37]. Moreover, g-SiC3 has semimetallic properties and it can be used as a gas sensor for air pollutants [39]. Furthermore, SiC7 siligraphene has a good photovoltaic applications [40], and can be used as a high capacity hydrogen storage material [41]. It shows superior structural, dynamical and thermal stability comparing to other types of siligraphene and it is a novel donor material with extraordinary sunlight absorption [42]. The structural and electronic properties of silicene-like SiX and SiX (X = B, C, N, Al, P) honeycomb lattices have been investigated [43]. Also, the planarity and non-planarity properties for g-SiCn and g-SiC (n = 3, 5, and 7) structures have been studied [44].

The excellent properties of siligraphene motivated us to study CSi7 and GeSi7, in order to find a new approach of silicon buckling and band gap control and to obtain new electronic and optical properties. Here we call CSi7 and GeSi7 siligraphene. We choose carbon and germanium atoms respectively for CSi7 and GeSi7 because these atoms, same as silicon atom, have four valence electrons in their highest energy orbitals. Using density functional theory, we show that both structures are stable but CSi7 is more stable than GeSi7. The carbon atom in CSi7 decreases the buckling, while germanium atom in GeSi7 increases the buckling. It is shown that CSi7 is a semiconductor with 0.24 eV indirect band gap [45] but GeSi7, similar to silicene, is a semimetal. Also, we investigate the effects of strain and we show that for CSi7, the compressive strain can increase the band gap and the tensile strain can decrease. At sufficient tensile strain (>3.7%), the band gap of CSi7 becomes zero and thus the semiconducting properties of this material change to metallic properties. As a result, the band gap of CSi7 can be tuned by strain and this...
material can be used in straintronic devices such as strain sensors and strain switches. For GeSi_7, strain does not have any significant effect on it. In contrast, GeSi_7 has high dielectric constant and can be used as a 2D material with high dielectric constant in advanced capacitors. Finally, we investigate the optical properties of these materials and we find that the light absorption of both CSi_7 and GeSi_7 are significantly greater than the light absorption of silicene. Because of high absorption of CSi_7 and GeSi_7, these materials can be considered as a good candidate for solar cell applications. It is worth to mention that germa-silicene, GeSi_7, is a new 2D material proposed and studied in this paper, while carbosilicene, CSi_7, has been proposed previously as a member of siligraphene but only its band structure has been studied [44 46]. The rest of the paper is organized as follows. In Sec. II, method of calculations is introduced and the results and discussion are given in Sec. III. Section IV contains a summary and conclusion.

II. METHOD OF CALCULATIONS

Density functional theory (DFT) calculations are performed using the projector-augmented wave pseudopotentials [47] as implemented in the Quantum-ESPRESSO code [48]. To describe the exchange-correlation functional, the generalized gradient approximation (GGA) of Perdew-Bruke-Ernzerhof (PBE) is used [49]. After optimization, the optimum value for the cutoff energy is obtained equal to 80 Ry. Also, Brillouin-zone integrations are performed using Monkhorst-Pack [50] and optimum reciprocal meshes of 12×12×1 are considered for calculations. At first, unit cells and atomic positions of both CSi_7 and GeSi_7 are optimized and then their electronic properties are determined by calculating the density of states and band structure. Moreover, their optical properties are determined by calculating the absorption and the imaginary and real parts of dielectric constant.

III. RESULTS AND DISCUSSION

A. Structural properties

FIG. 1. (a) Top view of silicene and (b) Si_8 unit cells. (c) Side view of Si_8 unit cell.

By increasing silicene unit cell [see Fig. 1(a)] in x and y direction twice, Si_8 has been constructed [see Fig. 1(b)] in hexagonal lattice (i.e., α = β = 90°, γ = 120°). In physical view, both silicene and Si_8 have the same physical properties because by increasing both unit cells, silicene monolayer has been achieved. In this work, Si_8 unit cell considered because CSi_7 and GeSi_7 can be constructed by replacing a silicone atom with a carbon or a germanium atom. After relaxation, the bond length of Si_8 was d = 2.4 Å [see Fig. 1(a)] and lattice parameters were |a| = |b| = 7.56 Å and |c| = 14.4 Å [see Figs. 1(b) and 1(c)] and buckling parameter Δ = 0.44 Å [see Fig. 1(c)] which has a good agreement with previous works [51 53]. Here c is the distance to make sure that there is no interaction between adjacent layers. For carbosilicene, CSi_7, unit cell construction, a silicon atom can be replaced with a carbon atom as shown in Fig. 2. Because of structural symmetry of CSi_7 monolayer (see Fig. 6), the position of impurity atom is not important, and our calculations also show the same ground state energy for all the eight possible impurity positions. After relaxation, optimum lattice parameters are obtained as |a| = 7.49 Å and |c| = 12.86 Å for CSi_7 unit cell. Fig. 2 shows this structure before and after relaxation. For a more detailed explanation, we labeled atoms in this figure. It is observed that Si-C bond length (i.e., d_2−4 = 1.896 Å) is shorter than Si-Si band length (i.e., d_1−2 = 2.317, d_1−3 = 2.217 Å) because of sp^2 hybridization. Also, unlike graphene, the hexagonal ring is not a regular hexagon due to the electronegativity difference between C and Si atoms [44].

FIG. 2. Top view of CSi_7 unit cell (a) before and (b) after relaxation. Carbon atom is shown by yellow sphere and silicon atoms by blue spheres.

Fig. 3 shows the side view of CSi_7 unit cell. After relaxation, the buckling parameter between atoms 1 and 3 (Δ_1−3) is 0.1 Å whereas this parameter for atoms 2 and 4 (Δ_2−4) is 0.39 Å. So, CSi_7 has a structure with two different buckling parameters and one can use the carbon atoms to decrease buckling parameter of silicene. Silicene has one buckling and two sublattices [54], while carbosilicene has two bucklings and thus three sublattices including one for carbon atoms and two others for silicon atoms.

If we replace a silicon atom with a germanium atom as shown in Fig. 4 we could obtain germa-silicene, GeSi_7, structure. As we can see in this figure, the optimized parameters
are $|a|=|b|=7.8\,\text{Å}$, $|c|=11.98\,\text{Å}$ and the Si-Ge bond length and lattice constants are greater than those of Si-Si. Also, by comparing bond lengths and lattice parameters of GeSi$_7$ and CSi$_7$ structures, it is seen that the bond lengths and lattice parameters of GeSi$_7$ are significantly greater than those of CSi$_7$ which is due to the larger atomic number and thus atomic radius of germanium relative to the carbon.

The buckling parameters of germasilicene structure are depicted in Fig. 5. After relaxation, we find that the value of these parameters are $\Delta_{2-4} = 0.53\,\text{Å}$ and $\Delta_{1-3} = 0.43\,\text{Å}$. Therefore, GeSi$_7$ like CSi$_7$ has a structure with two different buckling and the germanium impurity atom increases the buckling of silicene. Bond length values and other structural parameters after relaxation are shown in Table 1.

We now introduce a new parameter for buckling as

$$\Delta_d = |\Delta_{2-4} - \Delta_{1-3}|$$

which shows the difference between two buckling parameters. Value of $\Delta_d$ for CSi$_7$ (i.e., 0.29 Å) is greater than that for GeSi$_7$ (i.e., 0.062 Å) which means the carbon impurity atom has a greater impact than germanium on siliconic buckling. This effect could be explained based on electronegativity difference. The electronegativity by Pauling scale is 2.55 [57, 58], 1.9 [59] and 2.01 [60] for carbon, silicon, and germanium respectively. Therefore, electronegativity difference is 0.65 for CSi$_7$ and 0.11 for GeSi$_7$ which show that CSi$_7$ has a greater electronegativity difference which leads to the in-plane hybridized bondings and reduces the buckling in comparison to the other cases.

Fig. 6 shows the charge density of a monolayer of CSi$_7$ and GeSi$_7$. The charge density of a monolayer of Si is also shown in this figure for comparison [see Fig. 6(a)]. The high charge density around the carbon and germanium impurity atoms [see Figs. 6(b) and 6(c)] shows charge transfer from silicon atoms to impurity atoms. Also, the electron aggregation around impurity atoms indicates ionic-covalent bonds in CSi$_7$ and GeSi$_7$ structures because of electronegativity difference.

Now, we calculate the cohesive and formation energies for these structures. The cohesive energy is $-4.81\,\text{eV/atom}$ and $-4.32\,\text{eV/atom}$ for CSi$_7$ and GeSi$_7$, respectively. The negative value of cohesive energy for CSi$_7$ and GeSi$_7$ means that these structures will not be decomposed into their atoms. The more negative cohesive energy, the more stable structure, so CSi$_7$ is more stable than GeSi$_7$. Also, the calculated cohesive energy for silicene is $-4.89\,\text{eV/atom}$ which is in good agreement with previous studies [59, 60] and shows CSi$_7$ has a stable structure with cohesive energy very close to silicene. Our calculations show the formation energy for CSi$_7$ and GeSi$_7$ structures are
TABLE I. Optimum lattice parameters $|a|$, $|b|$ and $|c|$, bond lengths $d_{1-2}$, $d_{2-4}$ and $d_{1-3}$ and buckling parameters $\Delta_{2-4}$, $\Delta_{1-3}$ and $\Delta_d$. All values are in Angstrom.

|     | $|a|$ | $|b|$ | $|c|$ | $d_{1-2}$ | $d_{2-4}$ | $d_{1-3}$ | $\Delta_{2-4}$ | $\Delta_{1-3}$ | $\Delta_d$ |
|-----|------|------|------|----------|----------|----------|----------------|----------------|----------|
| Si$_8$ | 7.65 | 14.4 | 2.4  | 2.4      | 2.4      | 0.44     | 0.44           | 0.44           | 0.44     |
| CSi$_7$| 7.49 | 12.86| 2.317| 1.896    | 2.217    | 0.1      | 0.39           | 0.29           | 0.29     |
| GeSi$_7$| 7.8  | 11.98| 2.287| 2.34     | 2.297    | 0.53     | 0.45           | 0.1           | 0.1      |

$0.16$ eV/atom and $-0.005$ eV/atom, respectively. So, the formation of CSi$_7$ (GeSi$_7$) from their constituents is endothermic (exothermic) because of the positive (negative) value of formation energy. On the other hand, positive formation energy for CSi$_7$ represents a high stability of this structure, while the negative or nearly zero value for GeSi$_7$ is attributed mostly to the high reactivity related to silicene.

B. Electronic properties

To investigate electronic properties of CSi$_7$ and GeSi$_7$, at first, we compare band structure of silicene, CSi$_7$ and GeSi$_7$ monolayers and we show the results in Fig. 7. As we can see in this figure, like graphene and silicene, GeSi$_7$ is semi-metal (or zero-gap semiconductor) with Dirac cone in point $K$. This is because the $\pi$ and $\pi^*$ bands cross linearly at the Fermi energy $E_F$. These band structures indicate that the charge carrier in silicene and GeSi$_7$ behave like massless Dirac fermions.

In contrast with GeSi$_7$, CSi$_7$ is a semiconductor with indirect band gap. The value of its indirect band gap is $0.24$ eV in $K - L$ direction which significantly less than its direct band gap value (i.e., $0.5$ eV in $K - K$ direction).

For a better comparison, an enlarged band structure of silicene and GeSi$_7$ is shown in Fig. 8. It is seen that, in point $K$, silicene and GeSi$_7$ have similar band structures with zero band gap, whereas CSi$_7$ has a band gap. In Dirac cone of graphene and silicene, $\pi$ and $\pi^*$ bands are made from the same atoms but these bands in GeSi$_7$ are made from two different atoms. To determine the Fermi velocity, $v_F$, the graphs for silicene and GeSi$_7$ must be fitted linearly near the Fermi level by using equation $E_{F+K} = \gamma k$. Then the Fermi velocity is given by $v_F = \gamma / h$. Our calculations show that $v_F$ is $5 \times 10^5$ m/s for silicene (which shows a good agreement with previous works) and $4.8 \times 10^5$ m/s for GeSi$_7$. A comparison between Fermi velocity in silicene and GeSi$_7$ indicates that Ge atoms in GeSi$_7$ do not have a significant effect on Fermi velocity. The total density of states (DOS) is also shown in Fig. 8. It is observed that the total DOS has a good agreement with the band structure.

We now investigate the effect of strain on the band structure of CSi$_7$ and GeSi$_7$ and the results are shown in Fig. 9. As we can see in Figs. 9(a) and 2(b), compressive strain has important effects on band structure of CSi$_7$ but it has no significant effect on GeSi$_7$ [compare these figures with Figs. 2(b) and 2(c)]. In the presence of compressive strain for CSi$_7$, both direct and indirect band gaps increase, respectively from $0.5$ eV and $0.24$ eV to $0.52$ eV and $0.44$ eV. But for GeSi$_7$, the zero-band gap remains unchanged and compressive strain cannot open any band gap. Fig. 9(c) shows the direct and indirect band gap variations of CSi$_7$ versus the both compressive and tensile strains. It is observed that both direct and indirect band gaps increase with increasing the compressive strain, while they decrease with increasing the tensile strain. The variation of band gaps versus strain $S$ is nearly linear and could be formulated by $E_F = -0.017S + 0.447$ for direct band gap and $E_F = -0.059S + 0.227$ for indirect one. Under strain and without strain, the direct band gap has significantly larger values relative to indirect band gap, thus it has no important effect on electronic transport properties in CSi$_7$. In contrast with GeSi$_7$, the strain is an important factor for tuning of band gap in CSi$_7$. For example, when the tensile strain increases above the band gap of CSi$_7$ disappears and this 2D material becomes a metal [see Fig. 9(c)]. This property of CSi$_7$ is
important in straintronic devices such as strain switches and strain sensors.

The complex dielectric function $\varepsilon = \varepsilon_r + i\varepsilon_i$ can be calculated for both polarizations of light: (i) parallel (x direction) and (ii) perpendicular (z direction), where $\varepsilon_r$ is the real part and $\varepsilon_i$ is the imaginary part of the dielectric function. This function is an important parameter for calculation of optical properties of materials. For instance, the real and imaginary parts of refractive index (i.e., $n = n_r + i n_i$) can be written as:

$$n_r = \sqrt{\frac{\left(\varepsilon_r^2 + \varepsilon_i^2\right)^{1/2} - \varepsilon_r}{2}}$$  \hspace{1cm} (2)$$

and

$$n_i = \sqrt{\frac{\left(\varepsilon_r^2 + \varepsilon_i^2\right)^{1/2} - \varepsilon_r}{2}}$$  \hspace{1cm} (3)$$

respectively. The absorption coefficient $\alpha$ is given by:

$$\alpha = \frac{2\omega n_i}{C}$$  \hspace{1cm} (4)$$

where $C$ is the speed of light in vacuum. The real parts of dielectric function of CSi$_7$, GeSi$_7$ and silicene are depicted in Fig. 10 for x and z directions. This figure shows that $\varepsilon_r$ in both directions are inhomogeneous because the graphs of $\varepsilon_r$ are not similar for the two directions. The root of real part (where $\varepsilon_r = 0$) represents the plasma energy (frequency) which for these materials it locates at 4.3 eV (1.04 $\mu$Hz) for x-direction. It can be seen from Figs. 10 a) and 10 b) that the values of static dielectric constant (the value of dielectric function real part at zero frequency or zero energy) in the x-direction are 12.3 for silicene and CSi$_7$ and 30 for GeSi$_7$, and in the z-direction are 2.4, 2 and 2.9 for silicene, CSi$_7$ and GeSi$_7$ respectively. Thus, for both directions GeSi$_7$ has the biggest static dielectric constant. Also, the static dielectric constant of GeSi$_7$ is significantly greater than graphene (1.25 for z-direction and 7.6 for x-direction). According to the energy density equation of capacitors (i.e., $E = \varepsilon_0 E_0 E^2 / 2$), by increasing dielectric constant $\varepsilon$, the energy density $u$ increases. Here, $E$ in the electric field inside the capacitor. So, materials with high dielectric constant have attracted a lot of attentions because of their potential applications in transistor gate, non-volatile ferroelectric memory and integral capacitors.

Among the 2D-materials, graphene has been used for electrochemical capacitors and supercapacitors. Since GeSi$_7$ has a high dielectric constant, it can be used as a 2D-material with high performance dielectric in advanced capacitors.

Fig. 11 shows absorption coefficient $\alpha$ for CSi$_7$ and GeSi$_7$. Absorption coefficient for silicene is also shown in this figure for comparison. The absorption coefficient shown in this figure for silicene is in agreement with previous works. There are two peaks for CSi$_7$: one locates in 1.18 eV (infrared region) and the other in 1.6 eV (visible region). The peak for silicene (at 1.83 eV) locates in visible region (1.8-3.1 eV). So, carbon atom increases and shifts the edge of absorption from the visible region to infrared region because it breaks the symmetry of silicene structure and it opens a narrow energy band gap in silicene band structure. For GeSi$_7$ there is an absorption peak in visible region (at 2.16 eV). Also, the peak height of GeSi$_7$ is larger than that of silicene and CSi$_7$.
FIG. 10. Comparison of real part of dielectric function for CSi$_7$ and GeSi$_7$ (a) in x direction and (b) in z direction. The graphs of silicene are also shown in this figure for comparison.

FIG. 11. Absorption coefficient for silicene, CSi$_7$, and GeSi$_7$.

On the other hand, sunlight radiation received by earth is comprising 5% ultraviolet, 45% infrared and 50% visible [77]. So, we investigate area under the absorption curve of CSi$_7$ and GeSi$_7$ in visible (from 1.8 to 3.1 eV), near infrared (from 1.13 to 1.55 eV) and infrared (<1.13 eV). Fig. 12 shows this area for silicene, CSi$_7$ and GeSi$_7$ in infrared, near infrared and visible spectrum regions. As we can see in this figure, the absorption of CSi$_7$ for all three spectrum regions and total absorption are significantly greater than those of silicene. The absorption of GeSi$_7$ is greater than that of silicene in infrared and visible regions and it is smaller in near infrared region, but the total absorption of GeSi$_7$ is significantly greater than the total absorption of silicene. For comparison, we also calculate the absorption coefficient in infrared region for siligraphene SiC$_7$, a new material studied recently [42]. The absorption for siligraphene in infrared region is equal to 2.7 which shows that CSi$_7$ with absorption 8.78 and GeSi$_7$ with absorption 6.31 have more than two times greater absorption relative to siligraphene in infrared region.

FIG. 12. Areas under the absorption curve for silicene, CSi$_7$ and GeSi$_7$ in infrared, near infrared and visible spectrum regions.

D. Summary and conclusion

We studied the structural, electronic and optical properties of CSi$_7$ and GeSi$_7$ structures using density functional theory within Quantum Espresso code. We showed that the carbon atom in CSi$_7$ decreases the buckling, whereas germanium atom in GeSi$_7$ increases the buckling which promises a new way to control the buckling in silicene-like structures. Both structures are stable but CSi$_7$ is more stable than GeSi$_7$. Band structure and DOS plots show CSi$_7$ is a semiconductor with 0.24 eV indirect band gap but GeSi$_7$, similar to silicene, is a semimetal. Strain does not have any significant effect on GeSi$_7$, but for CSi$_7$, the compressive strain can increase the band gap and tensile strain can decrease it. At sufficient tensile strain (> 3.7%), the band gap becomes zero or negative and thus the semiconducting properties of CSi$_7$ change to metallic properties. As a result, the band gap of CSi$_7$ could be changed and controlled by strain and this material can be used in straintronic devices such as strain sensor and strain switch. Furthermore, we investigated the optical properties of CSi$_7$ and GeSi$_7$ such as static dielectric constant and light absorption. The GeSi$_7$ has high dielectric constant relative to CSi$_7$, silicene and graphene and can be used as a 2D-material with high performance dielectric in advanced capacitors. The light absorption of CSi$_7$ for near infrared, infrared and visible regions and its total absorption are significantly greater than those of silicene. The absorption of GeSi$_7$ is greater than that of silicene in infrared and visible regions and it is smaller in near infrared region, but the total absorption of GeSi$_7$ is significantly greater than the total absorption of silicene. Because of high absorption of CSi$_7$ and GeSi$_7$, these materials can be considered as proper candidates to solar cell applications.
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