



First-principle analysis of photoelectric properties of silicon-carbon materials with graphene-like honeycomb structure



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ABSTRACT

This study uses the first-principle pseudo-potential method based on density functional theory to investigate the structure stability, geometrical structure, electronic structure, deformation charge density, atomic orbital and bond population, and optical properties of graphene-like structures $\text{Si}_x\text{C}_{1-x}$ ($x = 0.02, 0.08, 0.2, 0.4, 0.5$). Geometrical structure results show that the substituted Si atoms induce the structural deformation along the directions on the graphene plane without any buckling. The cohesion energy calculation indicates that these substituted systems are absolutely stable. The pure graphene forms a zero-band gap semiconductor, and the densities of states near the Fermi energy are composed mainly of C-2p. With Si doping, a direct band gap appears at the K point in the Brillouin zone. The band gap increases from 0.15 eV to 2.60 eV with the increase in Si content. The densities of states of $\text{Si}_{0.50}\text{C}_{0.50}$ near the Fermi energy are composed mainly of C-2p and Si-3p. After substituting Si atoms for C atoms, charges transfer from Si atoms to C atoms, the covalent bond of C–C weakens, and the symmetry of the covalent bond of graphene is destroyed, which is very important for the appearance of direct band gap at the K point. All curves of optical properties of $\text{Si}_x\text{C}_{1-x}$ ($x = 0.02, 0.08, 0.2, 0.4, 0.5$) move to the high energy with the increase in Si content. Among those of all the studied $\text{Si}_x\text{C}_{1-x}$ systems, $\text{Si}_{0.50}\text{C}_{0.50}$ obtains the lowest static dielectric constant of 1.60, the lowest refractive index of 1.26, the highest absorption coefficient of $6.79 \times 10^4 \text{ cm}^{-1}$. The results will give a guide for design and application of the single layer SiC in further experimental and the theoretical investigations.

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1. Introduction

Two-dimensional materials are ideally suited for next-generation nano-electronic devices because their complex structures can be easily fabricated unlike those of one-dimensional materials. Among these materials, graphene is the most widely studied [1,2]. This material is a discovered form of carbon [1] that consists of only one plain layer of atoms arranged in a honeycomb lattice, and exhibits several interesting properties [3–10]. In the past few years, graphene has attracted considerable research attention and is becoming the leading edge and major concern in various relative fields. The structure and properties of modified graphene can be effectively modulated, thereby achieving abundant functions and applications. Graphene is used widely in micro-electronic devices because of its adjustable band gap, carrier

concentration, and carrier polarity. However, pristine graphene does not have a band gap, a property that is essential for many applications, including transistors. Thus, engineering the band structure of graphene becomes a research hotspot [11–18].

Doping is a common and important approach to tailor the electronic properties of semiconductor materials. In semiconducting carbon nanotubes (CNTs), substitutional B [19,20] and N atoms [20,21] act as acceptors or donors; they insert levels in the gap or cause a semiconductor–metal transition, thereby resulting in the p-type or n-type semiconducting behavior of CNTs, respectively [22,23]. Doping can also dramatically alter the electrical properties of graphene. Theoretical studies show that substitution doping can modulate the band structure of graphene. For example, Deifallah et al. [24] investigated the structure and electronic properties of single layers of carbon nitride graphenes. They found that carbon nitride graphenes form semiconductors with band gaps of up to 5 eV. Martins et al. [25] studied the electronic and transport properties of graphene nanoribbons (GNRs) doped with B atoms. They found that the substitutional B atoms at the nanoribbon

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edges suppress the metallic bands near the Fermi level, thereby forming a semiconducting system. Cervantes-Sodi et al. [26] and Lherbier et al. [27] found that N or B substitutions cause semiconductor–metal transitions in graphene. Wei et al. [28] demonstrated the feasibility of synthetic N-doped graphene by experimentation. Wang et al. [29] fabricated an n-type graphene field-effect transistor that operates at room temperature by doping N atoms to edge-terminated GNRs. The SiC nanotubes with an atom ratio of 1:1 have been synthesized via substituting C atoms in the CNTs with Si atoms in 2002 [30]. Substituting C atoms in graphene with Si atoms may obtain monolayer SiC of tunable band gap and optical properties. Only Zhou et al. [31,32] calculated the structural and electronic properties of Si-substituted graphene. However, the optical properties of silicon-carbon materials with graphene-like honeycomb structure have not been theoretically and empirically investigated.

This study investigates a series of calculations of Si-substituted graphene. Notably, the band gap and optical properties of this material can be tuned using different substitutional concentrations.

2. Computational models and details

Density functional theory calculations are performed using the CASTEP computer program [33]. In the geometry optimization, electronic structure and optical properties calculations, the electron exchange and correlation are obtained by using the generalized gradient approximation (GGA) with the interpolation formula of Perdew, Burke, and Ernzerhof (PBE) [34]. The electron–ion interaction is described by an ultra-soft potential generated using the optimization scheme of Vanderbilt [35]. The valence electronic wave functions are expanded in a plane-wave basis set up to an energy cutoff of 380 eV. Accordingly, the total energy of the unit cell is converged to 1.0×10^{-6} eV/atom. The Brillouin zone integration is performed using a $4 \times 4 \times 1$ Monkhorst–Pack set [36]. The atomic position and size of systems are relaxed. A $5 \times 5 \times 1$ supercell consisting of 50 C atoms are used for Si substitution with the concentrations of 2%, 8%, 20%, 40%, and 50%. Fig. 1 shows the structure geometrical models for Si substitution at different concentrations (The little gray balls are C atoms, and the big yellow¹ balls are Si atoms).

To check the stability of Si substitution on graphene, the cohesion energies of these substituted systems are calculated. The cohesion energy formula is [37] $E_{coh} = (E_{tot} - E_{atom}^A - E_{atom}^B)/N$, where E_{coh} is the cohesion energy; E_{tot} is the total energy of the system; E_{atom}^A and E_{atom}^B are the total energies of free atom A and B, respectively; and N is the total atom number in the system.

Table 1 shows the cohesion energies of these substituted systems.

As compared with the value of -7.9 eV/atom in pure graphene [38], the values for these Si_xC_{1-x} structures show good stabilities.

3. Results and discussion

3.1. Geometry structure

Fig. 2 shows the relaxed structure of Si_xC_{1-x} ($x = 0, 0.02, 0.08, 0.2, 0.4, 0.5$). As shown in the Fig. 2, with substituting C atoms by Si, more and more C–Si bond formed in the Si-substituted systems. Because of the bond length of C–Si is larger than that of C–C, when C atoms substituted by Si, the bond angles of C–C–C and C–Si–C near the Si are not 120° and the regular hexagon honeycomb struc-

ture exhibits a slight deformation at the plane direction. When the content of Si is 50%, all of the C–C bonds convert into C–Si bonds, and the bond angles of C–Si–C and Si–C–Si are all much closed to 120° and a quasi-honeycomb structure formed. Moreover, the graphene plane does not bend after substituting C atoms with Si. This result cannot be obtained by other metal atoms replacing C atoms in graphene [39].

Table 2 shows the optimized lattice constants of Si_xC_{1-x} ($x = 0, 0.02, 0.08, 0.2, 0.4, 0.5$) (Fixed axis C).

As shown in Table 2, when Si content is 2%, geometric deformation occurs near the Si atom. The lengths of C–Si and C–C bonds are 1.67 Å and 1.43 Å, respectively. The lattice constant of Si_xC_{1-x} ($x = 0, 0.02$) increases by approximately 1.14%, indicating graphene stretching. With the increase in Si content, the lattice constants of Si_xC_{1-x} and the average lengths of C–Si and C–C bonds all increase. All C–C bonds convert into C–Si bonds when Si content reaches 50%. The single-layer graphene completely converts into SiC single-layer structure, and the corresponding length of C–Si bond is 1.79 Å. Tensile graphene usually changes the angle between σ bond [40] and bond length. This phenomenon destroys the symmetry of graphene, thereby facilitating new equivalent sp^2 hybridization and changing the band structure finally.

3.2. Electronic structure

Band structure of graphene is calculated to check the accuracy of calculations. The zero-band gap structure, bond length, and bond angles conform to other previous calculations [32]. Fig. 3 shows the band structures of graphene and Si-substituted systems.

Fig. 3(a) shows that the contact point of valence band and conduction band is the Dirac point, which corresponds to the K point in the Brillouin zone. The Fermi energy is at the Dirac point, and the undoped graphene forms zero band gap semiconductors. C atom contains four valence electrons, with two electrons in the 2s and 2p layers. The two electrons in the 2s layer and the electron in the 2p layer form sp^2 hybridization, and a very strong σ bond forms between C atom. C–C links together to form the planar structure of honeycomb, and the distance of C–C is 1.42 Å. The remaining one electron in the 2p layer forms the $2p_z$ orbital, which is perpendicular to plane direction. The $2p_z$ orbital overlap results in a large π bond. σ bond is filled completely to form the deep valence band area, and the low physical effect is decided mainly by π bond.

As revealed from the partial density of states, the valence band and conduction band near the Fermi energy of graphene are composed of C-2p electrons. With the increase in Si content, several C–C bonds are replaced by C–Si bonds. The contribution of the electrons in the Si-3p gradually increases as the electrons in the C-2p gradually decrease. Notably, the conduction band becomes narrow. The band gap gradually increases from 0.15 eV to 2.60 eV with the increase in Si content from 2% to 50%. The top of valence band is formed by the electrons of C-2p, and the bottom of conduction band is formed by the electrons of Si-3p. Compared with other ionicity materials, such as GaN slice [41] (band gap 2.0 eV) and BN slice [42] (band gap 4.64 eV), the band gap of SiC slice is 2.60 eV after substituting half of C atoms in graphene with Si. This value is close to the previously reported band gaps of SiC slice of 2.50 [32] and 2.55 eV [43].

3.3. Charge density difference and population analysis

Charge density difference is the difference of the charge density after bonding and the corresponding atomic charge density. This difference is expressed as $\Delta\rho = \rho_{ABSC} - \rho_{ABNSC}$, where ρ_{ABSC} is the charge density after bonding; and ρ_{ABNSC} is the charge density before bonding.

¹ For interpretation of color in Figs. 1 and 4, the reader is referred to the web version of this article.

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