High-order nonlinear mechanical properties of g-SiC

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ABSTRACT

Silicon carbide is one of the most important semiconductors with wide bandgaps and various applications including power electronics, nuclear fuel particles, hostile-environment electronics, and blue light emitting diodes. We investigate the nonlinear mechanical properties of a proposed graphene-like planar hexagonal silicon carbide (g-SiC) monolayer using first-principles calculations. The strength of g-SiC is about half that of graphene. The ultimate strain of g-SiC is 0.2, 0.25, and 0.19, in the direction of armchair, zigzag, and biaxial, respectively. The Poisson’s ratio is 1.75 times that of graphene. In the nonlinear elasticity regime, we obtain the high order elastic constants up to fifth order. The stiffness monotonically increases with pressure, has the same trend as that of second order elastic constants but opposes to that of Poisson’s ratio. There is a minimum at \(-4\) GPa in the speed-pressure curve of compressive sound wave, different from the monotonic increment of shear waves. These theoretical mechanical properties provide elasticity limits for various applications of g-SiC.

1. Introduction

With a wide electronic bandgap (\(>3\) eV), silicon carbide (SiC) is one of the most important semiconductors. Cubic SiC possesses outstanding physical properties, including high hardness and Young’s modulus (\(~450\) GPa), high thermal shock resistance, and high thermal conductivity (Morkoç et al., 1994). In addition, SiC is chemically inert. SiC has been widely used for power electronics, sensors, blue light emitting diodes, composites, hostile-environment electronics, and heterogeneous catalyst supports (Ziegler et al., 1983; Falk et al., 2013; Pedersen et al., 2012; Wright et al., 2008; Oliveros et al., 2013; Ishikawa et al., 1998; Ivevovitch et al., 2013), as well as the diffusion barriers in tri-structural isotropic particle fuel (TRISO) (Chen et al., 2018, 2020; Peng et al., 2019). Besides bulk structure, nano SiC materials attract extensive attention because they are expected to behave differently due to quantum confinement and shape effects (Peng and De, 2014).

The graphene-like silicon carbide monolayer (i.e. g-SiC) is a single-atom-thick planar 2D material with hexagonal lattice, as shown in Fig. 1. It has been predicted to be stable with indirect band gaps of \(~2.55\) eV (Şahin et al., 2009; Hoat et al., 2020). As a result, g-SiC has promising applications in energy conversion and optoelectronics, including blue and ultraviolet Light-emitting diodes, and photo-voltaic devices (Şahin et al., 2009; Lu et al., 2012a; Ganguly et al., 1991). It also has potential applications in composites, sensors, field-effect transistors(FET), field emitters, nano electromechanical devices, catalyst, super-capacitors, bio-imaging probes and wave absorber. Despite its importance, single-atom-thickness g-SiC monolayers have not been fabricated yet. Only the ultra-thin two dimensional SiC film have so far been fabricated (Lin, 2012). Even in the absence of its real world realization, extensive theoretical studies have been performed. These studies include thermodynamical stability, electronic structures, as well as electronic, magnetic, optical, optoelectronics, and catalytic properties of g-SiC (Wu et al., 2015; Lin et al., 2013; Hsueh et al., 2011; Lu et al., 2012b; Wu and Guo, 2007; Luo et al., 2018; Sun et al., 2008; Bekaroğlu et al., 2010; Zhou et al., 2013; Yang et al., 2015; Zhang et al., 2014; Lin et al., 2015; Yu et al., 2010; Guilhon et al., 2015; Wang et al., 2016; Guo et al., 2018; Islam et al., 2019). However, their mechanical properties so far have attracted much less attention. A comprehensive study on the structure of the 2D Si\(_{1-x}\)C\(_x\) monolayer was performed using cluster expansion and particle-swarm optimization approach (Shi et al., 2015). It was found that the g-SiC monolayer (\(x=0.5\)) is the most stable thermodynamically among over 300 structures. However, its nonlinear mechanical behavior has not yet been investigated.

The mechanical properties of g-SiC are fundamental for other properties and applications. However, they have not been well studied. For example, the strain-engineering (tuning material properties by applying strain) of the indirect-direct band gap transition of g-SiC is foreshone (Lu et al., 2012a) but the mechanical stability is lacking. Recently, controlling electronic and optical properties of multi-layer g-SiC sheets were achieved by applying strain (Xu et al., 2016). However, it is unclear whether or not the g-SiC monolayers can sustain such a large deformation. Non-monotonic strain dependence of lattice thermal conductivity in monolayer SiC has been investigated (Guo et al., 2018).
The nonlinear mechanical properties and mechanical stabilities of g-SiC are unknown yet. The mechanical properties of nanomaterials form the basis for the development of mechanically responsive electronic devices (Ariga et al., 2012; Dai et al., 2015; 2016). Previous studies show that 2D monolayers including graphene (Peng et al., 2013a), g-BNC (Peng et al., 2013a), g-GeC (Peng et al., 2013e), g-InN (Peng et al., 2017) can sustain a large elastic deformation before reaching ultimate strain and then softens until fracture. We aim to examine if g-SiC monolayers have a similar behavior. The linear elastic properties and second-order elastic constants of g-SiC monolayers have been investigated (Wang et al., 2018). However, the higher order ($O(n > 2)$) elastic constants were unexplored. The importance of the higher order elastic constants is manifested by many applications related to anharmonicity (Hikt, 1981), including the nonlinear elasticity, phase transitions, lattice defects, phonon-phonon interactions, harmonic generation, temperature dependence of elastic constants, strain softening, thermal expansion, and echo phenomena (Peng et al., 2013g). For example, third-order elastic constants are important in tuning acoustic wave velocities due to finite-strain. It has also been reported that using local strain, nano devices including nano waveguides and surface acoustic wave sensors can be synthesized (Peng et al., 2012d; 2012a; 2015b).

The goal of this work is to assess the nonlinear mechanical behaviors of g-SiC for large strains using first-principles calculations. We computed the fifth order elastic constants, which are further linked with an accurate nonlinear elastic description. The pressure dependence properties are envisioned, including sound wave speed, stiffness, and second order elastic constants.

2. Methods

2.1. Elasticity theory

From elasticity theory (Crisfield, 1991), for a three dimensional isotropic material, when a macroscopically homogeneous deformation with deformation gradient tensor $F$ is applied, the lattice vectors of the deformed system are $h = FH$. The Lagrangian strain (Brugger, 1964) is defined as $\varepsilon = \frac{1}{2}(F^T F - I)$, where $I$ is the identity tensor. The strain energy density has a functional form of $\Phi = \Phi(\varepsilon)$, where $\varepsilon$ is the deformation gradient tensor. The elastic properties of the material are determined from $\Phi$, which is quadratic in strain for a linear elastic material. Nonlinear elastic constitutive behavior is established by expanding $\Phi$ in a Taylor series in terms of powers of strain $\varepsilon$. The symmetric second Piola-Kirchhoff stress tensor, $\Sigma_{ij}$, can be expressed (up to fifth order) as (Wei et al., 2009):

$$\Sigma_{ij} = \frac{\partial \Phi}{\partial \varepsilon_{ij}} = C_{ijkl} \varepsilon_{kl} + \frac{1}{2!} C_{ijklm} \varepsilon_{km} \varepsilon_{lm} + \frac{1}{3!} C_{ijklmnop} \varepsilon_{op} \varepsilon_{mn} \varepsilon_{pq} + \frac{1}{4!} C_{ijklmnopqr} \varepsilon_{op} \varepsilon_{mn} \varepsilon_{pq} \varepsilon_{sr} \varepsilon_{tq}. \quad (1)$$

where $\varepsilon_{ij}$ is Lagrangian elastic strain. Summation convention is employed for repeated indices; lower case subscripts range from 1 to 3. Herein $C$ denotes each higher-order elastic modulus tensor; the rank of each tensor corresponds to the number of subscripts.

The second-order elastic constants (SOEC), $C_{ijkl}$, third-order elastic constants (TOEC), $C_{ijklm}$, fourth-order elastic constants (FOEC), $C_{ijklmnop}$, and fifth-order elastic constants (FFOEC), $C_{ijklmnopqr}$, are given by the components of the fourth-, sixth-, eighth-, and tenth-rank tensors, respectively.

We used conventional Voigt notation (Nye, 1995) for subscripts: $1 \to 1, 22 \to 2, 33 \to 3, 23 \to 4, 31 \to 5$, and $12 \to 6$. It should be mentioned that for strain $\varepsilon_{ij} = 2\varepsilon_{22}, \varepsilon_{33} = 2\varepsilon_{33}, \varepsilon_{23} = 2\varepsilon_{12}$. Eq. (1) can be rewritten as

$$\Sigma_{ij} = \frac{\partial \Phi}{\partial \varepsilon_{ij}} = C_{ij} \varepsilon_{ij} + \frac{1}{2!} C_{IJK} \varepsilon_{J} \varepsilon_{K} + \frac{1}{3!} C_{IJKLM} \varepsilon_{J} \varepsilon_{K} \varepsilon_{L} \varepsilon_{M} + \frac{1}{4!} C_{IJKLMN} \varepsilon_{J} \varepsilon_{K} \varepsilon_{L} \varepsilon_{M} \varepsilon_{N} \varepsilon_{M}. \quad (2)$$

where the summation convention for upper case subscripts runs from 1 to 6. These constants are orientation-independent in g-SiC due to the six-fold rotation symmetry of the atomic lattice (Nye, 1995).

2.2. Nonlinear elasticity theory of 2D hexagonal lattice

The components of the elastic constants including TOEC, FOEC, and FFOEC tensors can be determined based on the symmetries of the honeycomb atomic lattice point group $D_6h$ which consists of a sixfold rotational axis and six mirror planes as formulated in ref (Peng et al., 2012b; Wei et al., 2009).

The fourteen independent elastic constants of g-SiC are determined by a least-square fit to stress-strain results from $ab initio$ DFT simulations in two steps. At the first step, we use least-squares fit to five stress-strain responses. Five relationships between stresses and strains is necessary because there are five independent FFOECs. We obtain the stress-strain relationships by simulating the following deformation states: uniaxial strain in the zigzag direction; uniaxial strain in the armchair direction; and, equibiaxial strain. From the first step, we find that the components of SOEC, TOEC, FOEC are over-determined (i.e., the number of linearly independent variables are greater than the number of constraints), and the FFOEC are well-determined (the number of linearly independent variables are equal to the number of constraints). Under such circumstance, the second step would be the least-square solution to these over- and well-determined linear equations.

2.3. Three deformation modes

At the first step, we carried out three deformations, uniaxial strain in the zigzag direction (case $a$), uniaxial strain in the armchair direction (case $c$) and equibiaxial strains (case $b$). For uniaxial strain in the zigzag direction, the strain tensor is,

$$\varepsilon^{z} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & \varepsilon_{zz} & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

where $\varepsilon_{zz}$ is the amount of strain in zigzag direction.
$N = \eta_a = \eta_c = \eta_i$ is the number of atoms in the unit cell. The energy per \( \eta \) neV in Ref. 6 for convenience, Peng et al. (2012c), Peng et al. (2013d), Peng et al. (2013c), De (2013a), Peng et al. (2015d), Peng et al. (2015e), Boron, 3. Results and analysis

Peng et al. (2015c).

Peng et al. (2013f), Peng et al. (2015a), Peng and De (2013b), Peng and Peng et al. (2013b), Peng et al. (2013e), Peng et al. (2013a), 2.4. Density functional theory calculations

The model consists of a 6-atom conventional unit cell with 3 carbon atoms and 3 silicon atoms. (Fig. 1). The reason that we chose this 6-atom conventional unit cell is to capture the soft mode in hexagonal rings (Marianetti and Yevick, 2010).

The energies of the system are computed using first-principles calculations within the frame of density-functional theory (DFT). All the DFT calculations were performed with the program of Vienna Ab-initio Simulation Package (VASP) (Kresse and Hafner, 1993). We adopted the generalized gradient approximations as parameterized by Perdew, Burke and Ernzerhof (PBE) for exchange-correlation functions (Perdew et al., 1996). The electrons included are the 2\( s^2 2p^2 \) and 3\( s^2 3p^2 \) electrons for carbon and silicon atoms, respectively with projector augmented wave (PAW) and pseudo-potential approach (Blöchl, 1994). The periodic boundary conditions are employed along all three directions. The calculations are performed at zero temperature for ground state. A plane-wave cutoff of 600 eV is used in all the calculations. The relaxation of the electronic degrees of freedom is achieved when the change of the total energy is smaller than 0.000001 eV in two consecutive steps. The atomic geometry was fully relaxed till the forces are less than 0.001 eV/Å. The irreducible Brillouin Zone of this 6-atom conventional cell is sampled with a Gamma-centered 22 22 1 k-mesh. To ensure that the inter-layer interaction is negligible, a 15 Å thick vacuum region is added. The second Piola-Kirchhoff (K-P) stress (Peng et al., 2012d) is used for the 2D forces per length with units of N/m.

The mechanical strains are applied to the system via three modes: uniaxial along the armchair direction, uniaxial along the zigzag direction, and biaxial, as described in subsection 2.3. Both tensile (strain > 0) and compressive strains are explored in all the three modes. The fourteen independent elastic constants of up to five-order elastic constants of g-SiC are determined by a least-squares fit to stress-strain results from first-principles calculations (Peng et al., 2012b). This method has been adopted to examine the mechanical properties of various graphene-like materials, including g-BN, g-GeC, g-GaN, g-TIN, g-ZnO, g-Boron, g-BNC, g-ZnS, g-MoS2, graphene-oxide, and g-InN.

Peng et al. (2012c), Peng et al. (2013d), Peng et al. (2013c), Peng et al. (2013b), Peng et al. (2013c), Peng et al. (2013a), Peng et al. (2013f), Peng et al. (2015a), Peng and De (2013b), Peng and De (2013a), Peng et al. (2015d), Peng et al. (2015c).

3. Results and analysis

3.1. Atomic structure

The lattice constant of g-SiC monolayers is the separation of the second nearest neighbor, or shortest C-C or Si-Si distance. The energies versus lattice constants were fitted with a fourth-order polynomial function, yielding the lattice constant of \( a = 3.096 \) Å. This value is in an excellent agreement with previous DFT studies (Şahin et al., 2009; Lu et al., 2012a; Shi et al., 2015), as well as the bulk counterpart 4H-SiC of \( a = 3.082 \) Å as measured in a Brillouin-scattering study (Kamitani et al., 1997). The bond length of C-Si bond is 1.787 Å, which is 0.367 Å (or 26%) longer than that in graphene. Our result of the bond length agree well with previous DFT calculations of 1.77 Å (Şahin et al., 2009) and 1.79 Å (Şahin et al., 2015). All atoms are coplanar. The C-Si-C and Si-C-Si angles are 120°. This result of atomic structure of g-SiC is in a good agreement with previous DFT calculations (Şahin et al., 2009; Lu et al., 2012a; Shi et al., 2015).

The cohesive energy (\( E_{coh} \)) per pair of Si-C atoms is calculated as

\[
E_{coh} = E_{[SiC]} - E_{[Si]} - E_{[C]}
\]

where \( E_{[SiC]} \) is the energy per Si-C pair in the g-SiC system, \( E_{[Si]} \) and \( E_{[C]} \) is the energy per silicon atom and carbon atom, respectively, in the isolated states of single atom, as calculated in a cubic simulation box with side-length of 15 Å. The \( E_{[Si]} \) is \(-0.088 \) eV and \( E_{[C]} \) is \(-0.083 \) eV. Our calculation of the cohesive energy of g-SiC is \(-13.883 \) eV. As a reference, previous DFT calculations report \( E_{coh} = 15.25 \) eV in Ref. Şahin et al. (2009) and \(-4.65 \) eV in Ref. Shi et al. (2015).

3.2. Strain energy profile

We examine the strain energy profiles for the assessment of the mechanical stability of g-SiC. All the atoms are allowed to relax in full degrees of freedom under the applied strain. We have studied both the compression and tension. The Lagrangian strains are used ranging from \(-0.1 \) to 0.4. The stepsize of 0.01 is adopted for all three deformation modes: armchair, zigzag, and biaxial deformation. We introduce the quantity of the reduced strain energy \( E_i = (E_{tot} - E_0)/n \) for convenience of comparison (Peng et al., 2017), where \( E_{tot} \) is the total energy of the strained system, \( E_0 \) is the total energy of the strain-free system, and \( n = 6 \) is the number of atoms in the unitcell. The energy profile of the system is the strain energy as a function of the strain as shown in Fig. 2. The energy profile can be used to measure the mechanical stability of a system.

\[
\text{armchair} \quad \text{zigzag} \quad \text{biaxial}
\]

(a)

(b)

Fig. 2. Energy landscape Reduced strain-energy strain responses of g-SiC (a) for the three deformation modes: uniaxial strain in armchair and zigzag directions, and equibiaxial strains, compared with that of graphene (b). Both tensile (\( \eta > 0 \)) and compressive (\( \eta < 0 \)) strains are applied. Both plots are in the same scale and same symbols for ease of view.
The $E_\sigma$ of g-SiC as a function of strain are illustrated in Fig. 2(a) for uniaxial armchair, uniaxial zigzag and equibiaxial deformations, compared to that of graphene (Fig. 2b). The upward opening of the strain energy curves in well shape ensures the positive curvature, and the Young’s modulus. Therefore, an upward opening implies the mechanical stability of the system at finite strains.

The shape of the strain energy profile also reveals asymmetry and isotropic properties. It can be seen that $E_\sigma$ is anisotropic for the three strain directions. $E_\sigma$ is non-symmetrical for compression ($\eta < 0$) and tension ($\eta > 0$). This non-symmetry suggests the anharmonicity (i.e., the deviation of a harmonic or quadratic behavior in response to disturbance) of the g-SiC structures when strain larger than 0.02. A harmonic region can be defined as the strains where the $E_\sigma$ is a quadratic function. Therefore, the harmonic region of g-SiC is defined by $-0.02 < \eta < 0.02$. The stresses increase linearly with an increment of the applied strains in this harmonic region. The corresponding anharmonic region is defined as the range of strains outside of the harmonic region. The high order elastic constants are essential in the anharmonic region. With even larger loading of strains, the system might fail in the plasticity region. The maximum strain in the anharmonic region is the critical strain (Peng et al., 2017), which, however, is not spotted in this study.

Compared to graphene (Fig. 2b), g-SiC has smaller strain energy at all the modes of deformation, indicating that the g-SiC is softer than graphene. Graphene shows abrupt deviation in armchair deformation at $\eta_u = 0.3$, implying the mechanical instability. The g-SiC monolayer does not show such deviation for strains up to 0.4.

### 3.3. Stress-strain curves

After strain energy, we investigate stress, the derivative of the strain energy with respect to strain. We use the second P-K stress to quantify the 2D system’s mechanical behavior. The second P-K stress versus Lagrangian strain relationships of g-SiC are shown in Fig. 3 for three modes, which are uniaxial strains along the armchair (Fig. 3a) and zigzag directions (Fig. 3b) and biaxial strains (Fig. 3c), compared to that of graphene in Fig. 3d-f, respectively. The ultimate tensile strength, or simply the ultimate strength, is defined as the maximum stress that a material can withstand in a tensile test. The corresponding strain is the ultimate strain. The systems of pristine g-SiC under strains beyond the ultimate strains is known as a meta-stable state. It can be destroyed by wave perturbations, defects, as well as high temperature effects including vibrations (Topsakal et al., 2010). The ultimate strain is in general determined by the intrinsic bonding strengths. The ultimate strain serves as an upper limit of elastic stability. Therefore, it is a key parameter for strain engineering and applications.

The ultimate strengths and strains are extracted from the stress-strain curves, as summarized in Table 1 corresponding to the different strain conditions. These results are compared to that of graphene (Peng et al., 2013g) and silicene (Peng et al., 2013h). When the tensile strain increases, the C-Si bonds are stretched, eventually leading to rupture. The bonds of those parallel to the direction of deformation are remarkably more severely stretched when the strain is employed along the armchair direction. The ultimate strain in armchair deformation is 0.2, larger than that of graphene (0.19) and silicene (0.17). Under the zigzag deformation, the ultimate strain is 0.25, also larger than that of both graphene (0.23) and silicon (0.21). For the biaxial deformation of g-SiC, we obtained the ultimate strain as $\eta_u = 0.19$, where all the chemical bonds are observed to be ruptured. This value is larger than silicon (0.17) but less than graphene (0.23). Our results show that the g-SiC monolayer is more flexible than both graphene and silicon in axial deformation. However, it is less flexible than graphene in biaxial deformation. We might expect less rippling in g-SiC monolayers than that of graphene.

Compared to graphene, g-SiC has much smaller ultimate tensile strengths, about 1/3 of that of graphene, in all modes of deformation.

### Table 1: Mechanical strength

<table>
<thead>
<tr>
<th>g-SiC</th>
<th>silicene (Peng et al., 2013h)</th>
<th>Graphene (Peng et al., 2013g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Sigma_1$</td>
<td>16.5</td>
<td>6.0</td>
</tr>
<tr>
<td>$\xi_1$</td>
<td>0.2</td>
<td>0.17</td>
</tr>
<tr>
<td>$\Sigma_2$</td>
<td>16.7</td>
<td>5.9</td>
</tr>
<tr>
<td>$\xi_2$</td>
<td>0.25</td>
<td>0.21</td>
</tr>
<tr>
<td>$\Sigma_3$</td>
<td>16.2</td>
<td>6.2</td>
</tr>
<tr>
<td>$\xi_3$</td>
<td>0.19</td>
<td>0.17</td>
</tr>
</tbody>
</table>

#### Fig. 3. Stress-strain relationship

Stress-strain responses of g-SiC (left panels) under the three deformation modes of armchair (a), zigzag (b), and biaxial (c) strain, compared with that of graphene (right panels) in d, e, f, respectively. $\Sigma_1$, $\Sigma_2$, $\Sigma_3$ denote the $x$, $y$, and $z$ components of stress. "Cont" stands for the continuum mechanics. DFT calculations are denoted as "DFT". Both tensile ($\eta > 0$, red region) and compressive ($\eta < 0$, aqua region) strains are applied. The tensile region is divided into two parts: elastic region (light green) and unstable region (yellow). The elastic region has two subdivisions: linear elasticity (pink) and nonlinear elasticity (dark violet). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
3.4. High order \((O_n(n > 2))\) elastic constants

The elastic constants are essential parameters for continuum mechanics which are widely used in finite element models. It is critical to predict the mechanical behaviors and properties of materials at macroscale. We have examined normal \((O_n(n = 2))\) and high order \((O_n(n > 2))\) elastic constants our model, including the second order \((O_2)\) elastic constants (SOEC), third order \((O_3)\) elastic constants (TOEC), fourth order \((O_4)\) elastic constants (FOEC), and fifth order \((O_5)\) elastic constants (FFOEC). The linear elastic response of a system can be well described by the second order \(O_2\) elastic constants. These high order \((O_n(n > 2))\) elastic constants are indispensable to characterize the nonlinear elastic response of \(g\)-SiC. Our results for the elastic constants and the elastic modulus from the DFT calculations are presented in Table 2, along with that of graphene and silicene.

The in-plane Young’s modulus \(Y_i\) and Poisson’s ratio \(\nu\) are obtained as \(Y_i = (C_{11} - C_{12}/C_{11})\) and \(\nu = C_{12}/C_{11}\). Our results of in-plane stiffness of \(g\)-SiC is \(Y_i = 160.1\) N/m, are much smaller than that of graphene (340.8 N/m) (Peng et al., 2013g), but larger than that (63.8 N/m) of silicene (Peng et al., 2013h). Interestingly, it is even smaller than the mean value (202.3 N/m) of that of graphene and silicene. This weakening reflects the fact that the stiffness varies non-linearly with respect to the chemical bond stretching. The bond length in \(g\)-SiC is 1.787 Å, about 26 percent larger than that of graphene (1.42 Å). We could understand this behavior as follows. The reduced strength of \(g\)-SiC compared to that of graphene can be simply understood from the viewpoint of bond strength. The Si-C bond is longer and therefore weaker than the C-C bond. Both \(g\)-SiC and graphene have the same crystal space group. As a result, the homogeneous increment of bond length leads to a reduction in mechanical strength.

For convenience of comparison, we need to convert the stress unit to the standard units of GPa. However, the out-of-plane dimension is obscure in 2D material. As the following convention, we use the interlayer van der Waals distance of 3.75 Å from a previous DFT study (Xu et al., 2016) as the out-of-plane dimension. Therefore, our results of the in-plane stiffness is equivalently 427 GPa, larger than the bulk modulus (245 GPa) of its bulk counterpart (Kamitani et al., 1997). The Poisson’s ratio of \(g\)-SiC is \(\nu = 0.314\), about 1.76 times of that of graphene, but similar to that of silicene.

The importance of higher order elastic constants is reflected by the practical needs because it is more convenient to apply pressure in experiments than chemical approaches. Therefore, it is essential to have the full stress-strain relationship with higher order elastic constants. Explicitly, we can obtain the pressure dependent second-order elastic moduli from the third order elastic description (Peng et al., 2012b). Another fact is that the higher order elastic constants play a key role in understanding the stress-strain behaviors. Stress-strain curves show that when the strain is larger than the ultimate strain the stress becomes softening. The fundamental mechanism of this softening is that the bond weakens and breaks under large strains from the view of electron bonding. In continuum mechanics, this softening behavior is determined by the TOECs and FFOECs. As a result, the negative values of TOECs and FFOECs play an essential role in the softening of \(g\)-SiC monolayer.

These fourteen non-zero components of the elastic constants (Table 2) can be classified into two groups. One is concerned with the normal terms, which are \(C_{11}, C_{22}, C_{111}, C_{222}\), and so on. The other is about shear terms, which are \(C_{12}, C_{112}, C_{1112}\), and so on. The strength of the material is determined by the normal terms while its flexibility is controlled by the shear terms. From the comparison of the data in the Table 2, one could conclude that the normal terms of \(g\)-SiC monolayers are smaller in magnitude than those of graphene. This result is consistent with the conclusion that the \(g\)-SiC is “softer” than graphene. This could be attributed to the weaker Si-C bonds in \(g\)-SiC comparing to C-C bonds in graphene. On the contrary, the shear terms do not show a clear trend. The \(g\)-SiC monolayers exhibit instability under large tension. It should be mentioned that the tensile strength could be an overestimate of instability (Li, 2012).

To illustrate the importance of the high order elastic constants, we investigate the components of nonlinear elasticity. At first, only the second order elasticity are considered, where the stress varies with strain linearly. As illustrated in Fig. 4 using the biaxial deformation as an example, the linear behaviors are valid within a small strain range, from \(-0.02\) to \(0.02\). The same conclusion can be drawn from the other deformation mode in Fig. 2.

Next, we consider the second and third order elastic constants. The

![Fig. 4. Order effect](image-url)
stress-strain curve can be accurately predicted within the range of \( \eta \leq \eta_f = 0.05 \) using the SOECs and TOECs. One step further, when we use the elastic constants up to the fourth order, the stress-strain curves from first-principles calculations can be well reproduced up to a strain as large as \( \eta_f = 0.1 \) as displayed in Fig. 4. Finally, for strains beyond 0.1, it is critical to include all elastic constants up to fifth order for an accurate modeling of the mechanical properties. The same collusion is achieved in the analysis of the uniaxial deformations.

It is worth noting that the higher order elastic constants can be determined experimentally (Brugger, 1965). Therefore, our theoretical investigations could trigger and serve as a guide for further experimental investigations.

3.5. Pressure effect on linear elastics

To demonstrate the importance and applications of the higher order elastic constants, we assess the pressure effect on the second-order elasticity moduli of g-SiC. For that purpose, we only need the third order elastic constants. Explicitly, when the in-plane pressure is introduced, the pressure dependent second-order elastic moduli \((C_{11}, C_{12}, C_{22})\) can be computed from \(C_{11}, C_{12}, C_{22}, C_{111}, C_{112}, C_{222}, Y_s, \) and \(\nu\) using the following relationships (Peng et al., 2012b):

\[
\begin{align*}
\tilde{C}_{11} &= C_{11} - (C_{111} + C_{112}) \frac{1 - \nu P}{Y_s}, \\
\tilde{C}_{12} &= C_{12} - (C_{112} + C_{122}) \frac{1 - \nu P}{Y_s}, \\
\tilde{C}_{22} &= C_{22} - (C_{122} + C_{222}) \frac{1 - \nu P}{Y_s}.
\end{align*}
\]

The results of the pressure effects are illustrated in Fig. 5. The second-order elastic moduli of g-SiC have a general trend to increase proportionally with an increment of the applied in-plane pressure. Furthermore, the Poisson’s ratio decreases nonlinearly as a function of in-plane pressure. It is worth noting that \(C_{11}\) is not symmetrical to \(C_{22}\) anymore. Only when \(P = 0\), \(\tilde{C}_{11} = \tilde{C}_{22} = C_{11}\). This anisotropy could be attributed to the anharmonicity of g-SiC under large strains.

3.6. Pressure effect on the sound wave speed

To further demonstrate the importance and applications of the higher order elastic constants, we investigate the anharmonic sound waves in g-SiC monolayer which could be tuned by the in-plane pressure, or biaxial strains. In practice, it is desirable to generate sound waves with different velocities depending via the deformation mode. In turn, sound waves also generate biaxial deformations as compressive or p-waves. Correspondingly, s-waves generate shear deformations. The speed of these two types of sound wave can be calculated from the elastic constants using the following equations:

\[
\begin{align*}
\rho_s \nu_s &= \frac{\tilde{Y}_s(1 - \nu)}{\rho_m(1 + \nu)(1 - 2\nu)}, \\
\rho_p \nu_p &= \frac{\tilde{C}_{11}}{\rho_m}.
\end{align*}
\]

The dependences of both \(\nu_p\) and \(\nu_s\) on in-plane pressures, or negative biaxial stress, are plotted in Fig. 6. Both \(\nu_p\) and \(\nu_s\) monotonically increase with an increment of the in-plane pressure. Therefore, by applying the biaxial strain, both the speed of p-waves and s-waves can be tuned.

Besides the speed along, the ratio of the two speeds, \(\nu_p/\nu_s\), is a key parameter in the mechanics of materials (Peng et al., 2012b). More interestingly, this quantity relates to the Poisson’s ratio through

\[
\frac{\nu_p}{\nu_s} = \frac{1}{\nu}(1 + \frac{\nu^2}{1 - 2\nu}).
\]

It is well demonstrated that the ratio of \(\nu_p/\nu_s\) monotonically increases with the increase of in-plane pressure as shown in Fig. 6.

It is worth pointing out that a sound wave speed gradient could be obtained by applying stress on a g-SiC monolayer. As a result, the refraction of sound wave can in turn redirect the propagation path of the waves. In this way, the sound wave can move following a curved path (Everest, 2001). It is a characteristic that the radius of curvature is inversely proportional to the speed gradient along the sound path. In addition, a negative strain gradient could result in a negative sound speed gradient. This sound velocity gradient then can be used to form a sound channel. As a result, this property could be used to build wave guides and surface acoustic wave (SAW) sensors (Benes et al., 1998; Weigel et al., 2002). Therefore, we propose that g-SiC-based nano-devices of SAW sensors, filters and wave guides can be synthesized using
local strains. With the advantage of avoiding extrinsic defects (Peng et al., 2019a), this new route could be beneficial to fabricate the new next generation electronics.

3.7. Elastic stability

Born elastic stability criteria are in general applied to 3D crystals (Mouhat and Coudert, 2014). For 2D crystal systems, the necessary and sufficient elastic stability conditions are that the strain energy density is always positive for any applied strain $\eta$ as (Mazziarz, 2019):

$$\Phi(\eta) > 0, \forall \eta \neq 0.$$  \hspace{1cm} (13)

In this point of view, our results of strain energy profiles illustrate the elastic stabilities of g-SiC monolayers as shown in Fig. 2(a).

In terms of elastic stiffness tensor, the necessary and sufficient elastic stability conditions for 2D hexagonal lattice are follows:

$$
\begin{cases}
C_{11} > 0; \\
C_{11} > |C_{12}|.
\end{cases}
$$  \hspace{1cm} (14)

Our results of the second order elastic constants $C_{11} = 177.6 \text{ N/m}$ and $C_{12} = 177.6 \text{ N/m}$ (Table 2) indicate the elastic stability according to the stiffness tensor criteria (Eq. (14)).

It is worth mentioning that in general there are temperature-induced instability in 2D materials, for example, in 2D transition metal oxides as reported in Ref (van Gog et al., 2019). In addition, there are temperature-induced phase-transition in 2D materials (Maity et al., 2018). Under high temperature, the 2D materials will decompose, see example in Ref. Wang et al. (2015). The thermal instabilities of 2D materials are an interesting research topic. However, such an investigation is not the focus of the present study.

4. Conclusions

We have carefully investigated the nonlinear mechanical properties and mechanical stabilities of g-SiC under large strains by means of first-principles calculations. We demonstrated that g-SiC exhibits a nonlinear elastic deformation up to an ultimate strain, which is 0.20, 0.25, and 0.19 for the three examined directions of armchair, zigzag, and biaxial directions, respectively. The deformation and failure behaviors, as well as the ultimate strength are anisotropic due to the anharmonicity, which require nonlinear elasticity. Compared to graphene and silicene, g-SiC possesses a relatively low in-plane stiffness (160.1 N/m) and a large Poisson ratio (0.314).

We emphasize the nonlinear elasticity of g-SiC monolayers that are formulated with higher order elastic constants. We therefore establish an accurate continuum description of the elasticity properties of g-SiC with up to fifth order elastic constants that consist of fourteen independent components. These 14 high order elastic constants are computed from the stress-strain curves obtained from DFT calculations. We have demonstrated the importance of the high order elastic constants with three applications of these high order elastic constants. First, we illustrated that the TOECs, FOECs, and FFOECs are indispensable for accurate modeling of the mechanical properties for strains larger than 0.02, 0.05, and 0.10, respectively. Second, the second order elastic constants including in-plane stiffness are predicted to monotonically increases with pressure. The Poisson’s ratio of g-SiC monotonically decreases with increasing pressure. At last, the sound velocity of compression wave has a minimum at in-plane pressure of ~4 GPa while that of shear wave monotonically increases with pressure. These high order elastic constants are key parameters in the development of a classical continuum mechanics model for large deformations in finite element analysis. Our results for the mechanical properties including the elastic limits provide a safe-guide for strain-engineering g-SiC based electronics.

CRediT authorship contribution statement

Qing Peng: Conceptualization, Methodology, Data curation, Visualization, Investigation, Formal analysis, Validation, Writing - original draft, Writing - review & editing.

Declaration of Competing Interest

None.

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Supplementary material

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