Thermal and tensile properties of diamondene at finite temperature: A molecular dynamics study

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HIGHLIGHTS

- Without being passivated, diamondene is not stable at room temperature. It is stable below 280 K.
- Diamondene has isotropic in-plane thermal expansion with coefficient of ~8.3×10⁻⁶/K and has orthotropic elasticity.
- When stretched in armchair direction at 250 K, bond breakage-induced shear band appears in diamondene if strain > 0.176.
- When stretched in the zigzag direction at 0.1 K, diamondene behaves elastic-to-brittle damage if strain > 0.368.

GRAPHICAL ABSTRACT
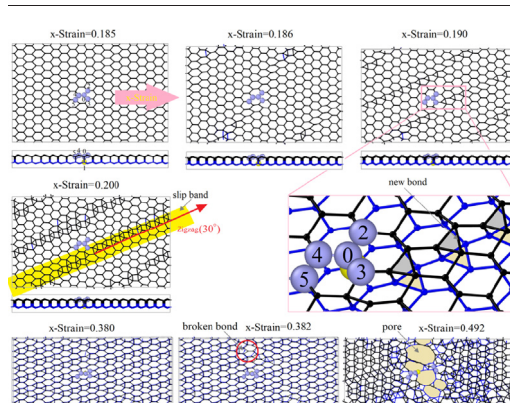
Diamodene under uni-axial stretching

ABSTRACT

Diamondene can be formed by compressing two or more layers of graphene. As a sp^2/sp^3 orbital hybridized two-dimensional material, its physical properties are different from either graphene or diamond. This new material is ultrahard on a SiC substrate. Here, we focus on the thermal stability and the basic mechanical properties of diamondene without passivation. We used molecular dynamics calculations to show that diamondene has isotropic thermal expansion at temperature below 280 K. At room temperature, it is unstable. When stretched along the armchair direction, diamondene has a maximum elastic strain that is higher than 0.17. It then experiences further plastic deformation before collapse. However, when stretched along the zigzag direction, the maximum elastic strain is higher than 0.3, e.g., 0.318 at 250 K. The ribbon collapses soon after the strain exceeds the critical value. Hence, diamondene has an elastic-brittle property along the zigzag direction. This excellent elasticity is significant for applications of diamondene in a flexible device.

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1. Introduction

Carbon has many allotropes due to its $2s^2 2p^2$ electron configuration, and the physical properties of carbon materials are dependent on their bond topology. These allotropes have various of configurations. For example, fullerene [1] is a 0-dimensional material. Made up of one-

dimensional material, carbon nanotubes [2] are popular in the design of nanodevices or other applications, e.g., yarn muscle [3–8]. Graphene was first reported over 10 years ago [9]. The current bulk production of graphene leads to a wide application of two-dimensional material in various fields [10–12]. Diamond is a typical 3-dimensional carbon material. Due to its super high hardness and excellent optical properties, it has been used in many engineering applications and in daily life. Besides the allotropes, artificial carbon materials [13–15] have also recently been developed.

Carbon materials with either the pure sp^2-sp^2 orbital hybridization (carbon nanotube and graphene) or with pure sp^3-sp^3 bond configuration (diamond) have long been known. More recently, people also have discovered new carbon allotropes with sp^2/sp^3 hybridization [16–20]. Of these, diamondene is formed by compressing two layers of graphene. Its physical properties are different from graphene or diamond. Barboza et al. [19] revealed the unique electrical properties of diamondene which behaves as a 2D ferromagnetic semiconductor with spin polarized bands. Recently, Martins et al. [20] reported the formation process of diamondene using a diamond anvil cell coupled with Raman. At pressures over 5 GPa, the inter-layer distance between the two sheets of graphene is reduced from 0.22 nm to 0.166 nm, resulting in diamondene. In other experiments, one sheet of graphene is passivated by hydrogen or hydroxyl groups, this influences the critical pressure for forming diamondene.

Gao et al. [21] tested the mechanical properties of the new two-dimensional materials on SiC substrate (0001), and they concluded that its in-plane stiffness and hardness are comparable to diamond. Cai et al. [22] studied the buckling behavior of nanotubes from diamondene. With both outstanding electrical and mechanical properties, diamondene is useful in various applications, e.g., micro-electromechanical systems, semiconductor components.

Here, we focus on the basic mechanical properties of diamondene without passivation (Fig. 1). We used a molecular dynamics approach to study the thermal expansion of diamondene at finite temperature. Uniaxial loading tests were performed to identify the in-plane mechanical properties of diamondene. We also studied the orientation dependence of the physical properties. The stability of the nanomaterial at finite temperature was evaluated.

2. Models and methodology

Here, molecular dynamics simulations were used to reveal the physical properties of diamondene as shown in Figs. 1 and 2. Calculations were performed in the open source code LAMMPS [23]. To calculate the mechanical properties of the pure carbon nanostructure via molecular dynamics, the Adaptive Intermolecular Reactive Empirical Bond Order (AIREBO) potential [24] was adopted to evaluate the interaction among neighbor atoms. The potential function has three items and reads,

$$P_{AIREBO} = P_{REBO} + P_{Torsion} + P_{L\text{-}J} \tag{1}$$

The first item on the right side of the equation can evaluate the bonding interaction with a cut-off value of 0.2 nm for carbon materials. This term considers the bond order with respect to bond generation or breakage. The second item describes various dihedral angle preferences. The local large rotation of the neighboring bonds can be described by this item. The third item reflects the nonbonding interactions, i.e., the 12-6 Lennard-Jones potential [25], between neighbor atoms with distance of <1.02 nm in this study. The AIREBO potential reflects the bonding and nonbonding interactions among atoms, and it is widely used in molecular dynamics studies on carbon nanomaterials or nanosystems [5, 20–22, 26].

Two models are involved in the molecular dynamics calculations to obtain the physical properties of diamondene. The model in Fig. 2a was used to calculate the geometric parameters and thermal expansion coefficients of diamondene at finite temperature. Two layers of diamondene were piled and relaxed at an NPT ensemble to calculate the thickness of diamondene. The other model (Fig. 2b) was used in uniaxial-tension tests to find the elasticity or damage behavior of diamondene. The system in a NPT ensemble is relaxed without pressure for 200 ps. During stretching, the strain ratio is of 0.001/ps with a time step of 1 fs. If we discuss the effect of strain ratio [27–30], then detailed values of strain ratios will be mentioned.

In each simulation, the first step is to build the nanoribbon. The bond lengths and bond angles are close to the ideal values. In the second step, potential minimization is fulfilled to reshape the geometry of the ribbon. In the third step, the ribbon is put in an NPT ensemble for
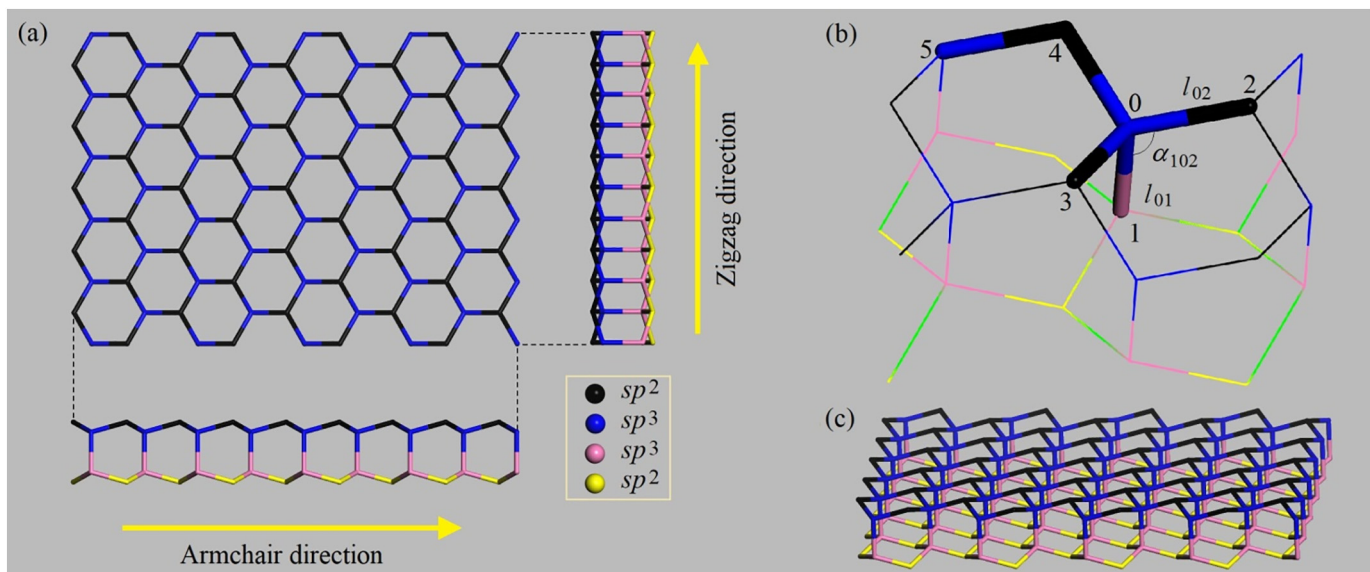
Fig. 1. Schematic geometry of diamondene. (a) Schematic of a diamondene ribbon with both side views along armchair and zigzag directions. (b) Representative geometry of diamondene with uniform in-plane deformation with two types (sp^2 and sp^3) of carbon atoms, 5 representative bond lengths, i.e., l_{01}, l_{02}, l_{03}, l_{04} and l_{45}, and 6 representative bond angles, i.e., α_{102}, α_{103}, α_{203}, α_{204}, α_{304} and α_{045}; l_{01} means the bond length between atom 0 and atom 1. The angle between bonds 01 and 02 is labeled as α_{102}. (c) Oblique-view of a diamondene ribbon.