

Metallic and ferromagnetic carbon allotrope discovered using molecular precursors

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Although carbon materials are known for their versatile structures and properties, a crystalline phase of carbon being both metallic and ferromagnetic is yet to be found. Here, using an organic molecular precursor 3,3-dimethyl-1-butene (C_6H_{12}) and a comprehensive theoretical study, we have discovered a metastable crystalline carbon phase that is both metallic and ferromagnetic under ambient conditions. The new carbon structures, produced by stacking puckered layers of 4,6-membered rings, are found to exhibit exotic electronic, magnetic, and mechanical properties originating from a special $sp^2 - sp^3$ bonding topology. These findings open the prospect of realizing the metallic and ferromagnetic carbon allotrope in practice.

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I. INTRODUCTION

Due to its flexible bonding characterized by sp^3 , sp^2 , and sp^1 hybridization of the s and p orbitals, carbon forms numerous compounds with properties intimately related to the specific structures. Since the discovery of C_{60} fullerene [1], one has witnessed the emergence of many crystalline and amorphous carbon materials [2–6]. More than 500 hypothetical carbon allotropes were collected in the Samara Carbon Allotrope Database (SACADA) [7–11] and more have been added recently [12,13]. There has been a constant search for metallic and magnetic carbon materials. The electrical conductivity ($2 - 3 \times 10^5$ S/m) of graphite in its basal plane comes from the two-dimensional (2D) graphene layer with zero density of states at the Fermi level. A similar case is found in the bundle of armchair nanotubes, where the conductivity is from the isolated 1D nanotube along the bundle axis [14]. A cubic phase of carbon formed under 3 Tera-Pascal (3×10^{12} Pa) pressure was reported to be metallic, however, loses its stability when pressure is removed [15]. More metallic carbon allotropes have been found in theory (e.g., in Refs. [7,16–18]), although some of them are not mechanically stable under ambient conditions [16,17]. None of these carbon allotropes are magnetic. Even among the hypothetical carbon allotropes, carbon magnetism is extremely rare. We screened all the carbon structures in the SACADA database [7–11]. Out

of a total of 522 3D carbon allotropes, 81 are sp^2 - sp^3 structures with 51 being metallic. Among all the sp^2 - sp^3 carbon allotropes, only four of them contain magnetic carbon sites in the structure with paramagnetic or antiferromagnetic ground states and no ferromagnetic ones [see Table S1 of Supplemental Material [19]]. Hydrogenated/functionalized graphene [20–22], defective graphene and its stacked nanostructures [23–25], and amorphous carbon structures [5,20–22] may become magnetic due to unsaturated bonds. Carbon radicals carrying net magnetic moments can be stabilized in local structures with negative Gaussian curvature [26,27], leading to ferromagnetic behavior in the amorphous schwarzites carbon foam [28].

Thus, crystalline metallic carbon with a ferromagnetic ground state is yet to be discovered. Such a material is expected to be metastable with both three- and four-coordinated carbon atoms. Metastable carbon materials exist when the interconversion barriers are high enough to prevent them from transitioning to globally more stable phases. Besides the well-known diamond, many other carbon materials, e.g., C_{60} , other fullerenes, graphdiyne, etc., are metastable. One way to synthesize a metastable carbon in experiments is to use selected molecular precursors in the reaction. For example, C_{20} fullerene was produced by using the precursor dodecahedrane ($C_{20}H_{20}$) that bears close resemblance to its cage structure [29]. Similarly, graphdiyne that has a much higher energy (0.76 eV/atom) than that of graphene, was formed by using the hexaethynylbenzene ($C_{18}H_6$) molecular precursor [30]. It is energetically preferable for the precursors to maintain some of their original bonding features during the formation of the carbon structures, especially if the kinetics are fast enough. Here, we extend such experimental approaches to structural search by building and relaxing assemblies of selected molecular precursor units rather than individual atoms. A 3D crystalline carbon is discovered that is both metallic

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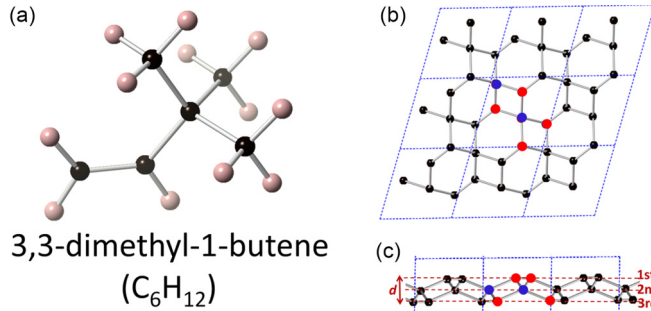


FIG. 1. (a) Structures of the molecular precursor 3,3-dimethyl-1-butene, (b) the top and (c) lateral views of U-graphene. The sp^2 and sp^3 bonded carbon atoms in the unit cell are in red and blue, respectively.

and ferromagnetic under ambient conditions. Further modeling and simulations show that the new allotrope and its related structures can be characterized by different stacking configurations of the puckered carbon layers. Each layer is a quasi-2D allotrope that consists of equal number of tetra- and hexamembered rings. We name these new carbon materials conductive ferromagnetic carbon (CFC). For simplicity of reference during the discussions, we will call them unusual carbon (UC) in this paper.

II. RESULTS AND DISCUSSION

The structure search for UC is based on orientationally assembled precursor units as detailed in Sec. 1 of Supplemental Material [19]. The method is in line with the random structure searching approach [31–34], where an ensemble of initial structures of the dehydrogenated 3,3-dimethyl-1-butene [Fig. 1(a)] units is chosen and each assembly is fully relaxed to the minimum energy using density functional theory (DFT) calculations. Detailed computational methods are given in Sec. 2 of Supplemental Material [19]. Based on the number of repeated structures (frequency of occurrence) found, the search is biased towards certain metastable structures that involve minimal configuration change in the precursor unit. The discovered UC structures are due to different

stacking configurations of a quasi-2D allotrope known as bex-tetrahexacarbon [35] characterized by equal number of tetra- and hexamembered rings [Fig. 1(b)]. As shown in Fig. 1(c), this particular quasi-2D allotrope referred to as U-graphene has a buckled structure with three equally separated ($d/2 = 0.65$ Å) sublayers of carbon. Its unit cell contains six carbon atoms with two sp^3 and four sp^2 carbon provided by the backbone structure of the precursor 3,3-dimethyl-1-butene (refer to Fig. M-1 and its discussion in the Supplemental Material [19]).

As shown in Fig. 2, the AA stacking of U-graphene results in a layered configuration termed UC-aa ($+0.75$ eV/carbon vs. diamond). The sp^2 carbon atoms exposed outside form a staggered arrangement between the neighboring layers. This leads to the interaction between the neighboring layers, originating from the p_z electrons of the sp^2 carbon. The interaction is weak in the AA stacking, with a small exfoliation energy of 0.27 J/m², which is like that of graphite (~ 0.30 J/m²). The AB stacking results in a sp^3 bonded configuration known as sodalite [36,37] termed UC-ab ($+0.38$ eV/carbon vs. diamond). The other configurations between the above two configurations include the $\frac{1}{2}$ AB stacking termed UC-aa' ($+0.70$ eV/carbon vs. diamond) and a mixture of $\frac{1}{2}$ AB and AB stacking termed UC-aa'ab ($+0.51$ eV/carbon vs. diamond). UC-aa' and UC-aa'ab structures have not been known before and can be formally named as 3,4-c trinodal and 3,4-c eptanodal, respectively, according to the naming methodology adopted in the Reticular Chemistry Structure Resource [11]. Also shown in Fig. 2 are the corresponding 3D views of these structures, with a tilted angle to illustrate the bonding between the layers and the arrangement of the carbon atoms in the direction perpendicular to the paper. Note that the visually triangular arrangement of carbon (numbered as 1, 2, and 3) in Fig. 2 is composed of three carbon atoms in different planes parallel to the paper. This special geometrical arrangement is due to the uniquely buckled structure of U-graphene and can lead to a local topology that can carry net magnetic moments as will be discussed later. Transitions between the structures can be realized by sliding the layers with respect to each other. As puckered atoms of one-layer approach another, new bonds can be formed as demonstrated by the

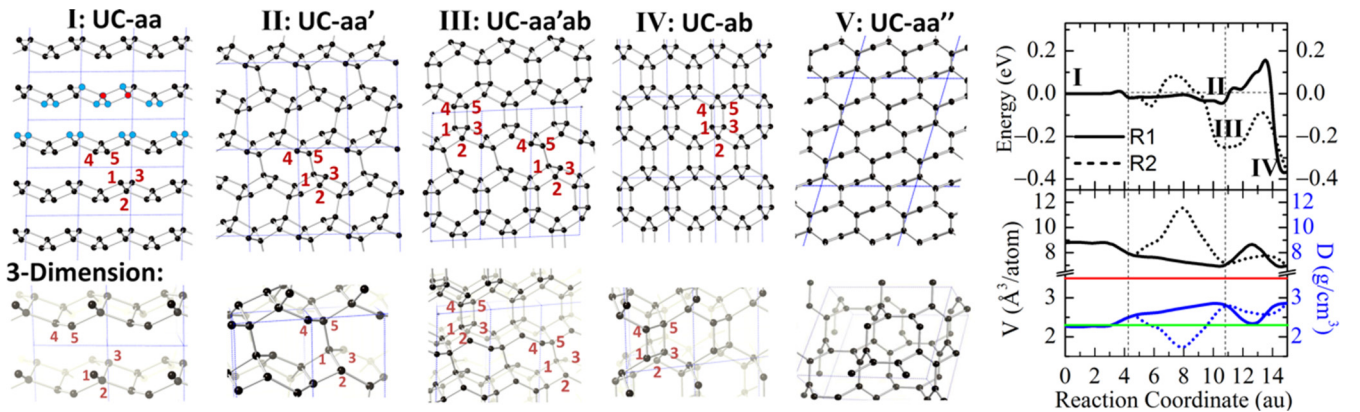


FIG. 2. UC structures (UC-aa, UC-aa'', UC-aa'ab, UC-ab, and UC-aa'') according to different stacking of U-graphene. Typical sp^2 and sp^3 sites are highlighted in blue and red in UC-aa, respectively. The corresponding 3D views show bonding between the layers and the arrangement of atoms. The calculated energy together with the volume (V) and density (D) profiles along the two reaction routes (R1 and R2) that connect these configurations are shown. The densities of graphite (green) and diamond (red) are shown for comparison.

connections between the numbered atoms (4 and 5 connecting to 1 and 3).

These structures are energetically related by continuous transitions, as shown by the calculated two reaction routes in Fig. 2 (also see Movies S1–S4 of Supplemental Material [19]), with UC-aa as the starting point and UC-ab as the end point. It is found, per Fig. S1 of Supplemental Material [19], that UC-aa' can undergo a phase transition below 600 K to a pure sp^2 bonded structure known as etb3-c [50] (termed UC-aa'' in Fig. 2, +0.42 eV/carbon vs. diamond). Although all these structures are found to be lattice dynamically stable (per Fig. S2 of Supplemental Material [19]), only UC-aa'ab, UC-ab, and UC-aa'' are protected from other phases by inter-conversion barriers higher than 0.4 eV/atom along the reaction route. Indeed, UC-aa'ab, UC-ab, and UC-aa'' are found to be kinetically stable above the room temperature, as shown in Figs. S3–S5 of Supplemental Material [19]. The symmetrized cells and lattice parameters of the carbon phases are given in Table S2 of Supplemental Material [19]. A full list of reflection peaks of the stable UC-aa'ab, UC-ab, and UC-aa'' is given in Table S3 of Supplemental Material [19]. The calculated Raman and IR active modes of the UC-ab and UC-aa'' structures are given in Table S4 of Supplemental Material [19]. The stable UC structures have higher densities than that of graphite (2.3 g/cm³) and lower densities than the density of diamond (3.5 g/cm³), per Fig. 2.

Notably, our structural search method based on the assemblies of orientated molecular units is the key to find the UC structures and identify their relation to the U-graphene. To prove this point, we used the particle swarm optimization [38] and conducted a regular structure search based on assemblies of individual carbon atoms, as put in Sec. 1 of Supplemental Material [19]. The only structure that can be found using such a method is UC-ab. It is not clear at all that the UC-ab structure is just one of the possible stacking configurations of the U-graphene layers without the other UC structures being identified by the search.

UC-aa'ab is found to be both metallic and ferromagnetic with a magnetic moment of 0.43 μ_B per unit cell in its ground state, as shown in Table I (refer to Sec. 2 of Supplemental Material [19] for the computation methods). According to the calculated electronic band structure in Fig. 3(a), parts of the valence bands of UC-aa'ab cut through the Fermi level, so that both electron and hole carriers contribute to the conductivity. With elevated temperature, electrons can be continually excited to fill the bands across the Fermi level and even further transfer to the bands hanging closely above. The origin of such electronic characteristics is due to those band edges contributed by the p_z orbitals from the sp^2 carbon atoms that are staggered between the layers (per Fig. 2). For instance, a tensile strain can pull away the p_z orbitals between layers, making the UC structure less conductive; while a compressive strain can enhance the conductivity by making the p_z orbitals more overlapping, as demonstrated in Fig. S6 of Supplemental Material [19].

The p_z electrons of the sp^2 carbon atoms are also the major magnetic bearers, as shown in Fig. 3(b). The origin of the carbon magnetism is due to the specific local topology in the UC-aa'ab structure, where each magnetic carbon [with the red arrow in Fig. 3(b)] is bonded to three fourfold

TABLE I. The number of magnetic carbon atoms (NMC) per unit cell, gained energy of the ferromagnetic ground state (ΔE_{FM}) in meV per unit cell (per magnetic carbon), and the magnetic moment (MM) in μ_B per unit cell (per magnetic carbon). The number of atoms per unit cell is given in the parentheses in the first column.

Structure	NMC	ΔE_{FM}	MM
1ab (UC-aa'ab) (18)	2	−9 (−4.5)	0.43 (0.215)
3 ab (30)	2	−6 (−4.5)	0.44 (0.22)
5 ab (42)	2	−7 (−3.5)	0.44 (0.22)
7 ab (54)	2	−7 (−3.5)	0.44 (0.22)
9 ab (66)	2	−12 (−6.0)	0.50 (0.25)
11 ab (78)	2	−11 (−5.5)	0.50 (0.25)
∞ ab (UC-ab) (12)	0	0	0
3 aa' (48)	8	−9 (−1.125)	1.28 (0.16)
5 aa' (36)	6	−1 (−0.167)	0.17 (0.028)
(1,3) ab (48)	4	−18 (−4.5)	0.92 (0.23)
(1,1,3) ab (66)	6	−28 (−4.67)	1.41 (0.235)

coordinated carbon atoms, leading to a frustration of tetravalent bonding and an unpaired spin. Without such local bonding, both UC-aa'' and UC-ab with pure sp^2 and sp^3 bonded carbon, respectively, are semiconducting and nonmagnetic, as shown by the calculated electronic structures in Fig. S7 of Supplemental Material [19]. The UC-aa'ab phase and especially its unique local structure of the magnetic carbon is found to be stable beyond room temperature, as shown by the molecular dynamics simulations (NpT) at high temperatures in Fig. S3 of Supplemental Material [19].

Note that, given the stacking nature of the UC structures, there can be an uncertainty in the number of layers to form the $\frac{1}{2}$ AB stacking or the AB stacking. The $\frac{1}{2}$ AB stacking is responsible for the local bonding structure of the ferromagnetic phase as in the UC-aa' and UC-aa'ab structures (Fig. 2), while the AB stacking is necessary to stabilize the structure at high temperatures as in the UC-aa'ab and UC-ab (Fig. 2) structures. With increasing number of AB-stacking layers in the unit cell as shown in Fig. 4(a), the resulting XRDs (e.g., “n ab” in Fig. 4(b) with $n = 3, 5, 7, 9, 11$) will change between

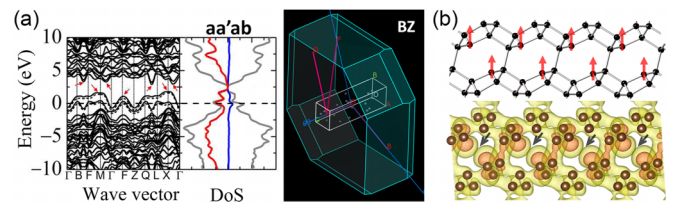


FIG. 3. (a) Calculated electronic structure of UC-aa'ab with the Brillouin zone (BZ) shown. The solid and dotted lines show the inequality of the two spins around the Fermi level (at zero). The gray lines in the density of states (DoS) correspond to spin-up and -down states, with the DoS of p electrons in red and the net spin at Fermi level in blue. The high symmetry k points are Γ (0,0,0), B(0.5,0,0), F(0,0.5,0), M(0.5,0.5,0), Z(0,0,0.5), Q(0,0.5,0.5), L(0.5,0,0.5), and X(0.5,0.5,0.5). (b) The primary magnetic sites in the ground state of UC-aa'ab are shown by red arrows in the top panel. Also shown is the calculated charge density of the ferromagnetic ground state, with the delocalized p_z electrons (bottom panel).

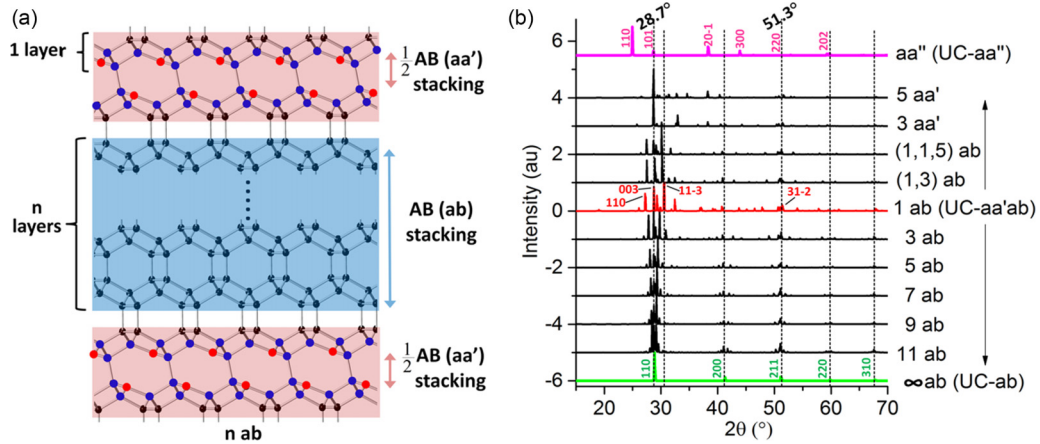


FIG. 4. (a) Structure models with increasing number of AB-stacking layers (in blue shades) separating each $\frac{1}{2}$ AB stacking (in red shades). The magnetic carbon sites are highlighted in red. (b) The XRDs of the structures due to different numbers of stacking layers are changing between the two extreme cases of U-aa'ab and U-ab.

the two extreme cases of UC-aa'ab [“1 ab” in Fig. 4(b)] and UC-ab [“ ∞ ab” in Fig. 4(b)]. The UC-aa'ab structure exhibits the largest number of peaks and UC-ab has the least number of peaks [Fig. 4(b)]. There will be relative peak shifting around 28° , 41° , 51° , 60° , and 68° into individual groups—from the many peaks in UC-aa'ab to only a few in UC-ab [Fig. 4(b)]. Interestingly, n can only assume odd numbers in these (n ab) structures to keep the bonding compatibility at the periodic boundary.

Simple combinations of the stacking disorder are also considered, including two $\frac{1}{2}$ AB-stacking layers separated by 1 and 3 AB-stacking layers in one case and three $\frac{1}{2}$ AB-stacking layers separated by 1, 1, and 3 AB-stacking layers in the other, as shown in Fig. S8(a) of Supplemental Material [19]. The corresponding XRDs of the two structures are similar to that of UC-aa'ab [Fig. 4(b)]. The disorder in the number of $\frac{1}{2}$ AB-stacking layers is further considered, including three $\frac{1}{2}$ AB-stacking layers separated by one AB-stacking layer in one case and five $\frac{1}{2}$ AB-stacking layers separated by one AB-stacking layer in the other, per Fig. S8(b) of Supplemental Material [19]. In these cases, there will be changes in the relative intensity of some peaks in the XRDs without obvious peak shift [Fig. 4(b)]. Thus, if randomized stacking happens in a UC sample, significant broadening around the five major peaks at 28° , 41° , 51° , 60° , and 68° are expected and the relative intensity of the peaks may change. However, per Fig. 4(b), the two peaks around 28.7° and 51.3° , respectively, corresponding to the (003) and (31 $\bar{2}$) crystal planes in the UC-aa'ab structure are seen in all cases, independent of the stacking disorder. We have further compared the simulated XRDs of all $sp^2 - sp^3$ carbon allotropes in the SACADA. None of them can simultaneously match these two peaks. Therefore, these XRD peaks could serve as the fingerprint to identify the UC phase in future experiments.

III. CONCLUSION

It is found that the ferromagnetism of UC is stable against the possible stacking disorder in the structure. As

shown in Table I, all the structure models above exhibit ferromagnetic ground states. In addition, increasing the number of AB-stacking layers in between the $\frac{1}{2}$ AB-stacking layers in the structure can enhance the stability of the ferromagnetic ground state and increase the net moment each magnetic carbon can carry. This is shown by the n ab and the (1,1,3) ab models in Table I, where the structures having the largest numbers of AB-stacking layers exhibit the lowest energy and largest magnetic moments per magnetic carbon. The estimated Curie temperature according to the energy gained by the ferromagnetic state relative to the paramagnetic state (28 meV/magnetic carbon, Table I) is about 340 K. Increasing the connected number of $\frac{1}{2}$ AB-stacking layers that contain the magnetic carbon will not stabilize the ferromagnetic state, as evidenced by the 3 aa' and 5 aa' models in Table I. Instead, the net magnetic moment carried by each magnetic carbon is significantly reduced and even demagnetized in the border layers (highlighted in green in the 5 aa' model) as shown in Fig. S8(b) of Supplemental Material [19].

Besides being metastable with significant interconversion barriers, these UC phases exhibit formation energies that are comparable or even lower than some synthesized carbon materials. Figure 5 shows the formation energies (relative to that of diamond) of the experimental realized carbon materials, including the C_{20} crystal [39], C_{60} crystal [40], cuboidal C_{60} [41], fullerene network [42], fullerite C_{24} [43], graphullerene [44], and fullertube C_{100} [45], in comparison with the formation energies of UC phases. Among these, most UC phases including the magnetic ones in Table I, such as the (1,1,3) ab phase, have lower formation energies than that of cuboidal C_{60} [41], with all the UC phases having formation energies lower than those of the C_{20} crystal [39] and fullerite C_{24} [43], but slightly higher than that of the recently synthesized graphullerene [44], as shown in Fig. 5(a). It is also found that, compared to these known carbon materials, the UC phases belong to a medium-energy carbon family with relatively high density closer to that of diamond, as shown in Fig. 5(b).

Interestingly, the structure of U-graphene is related to the structure of the pentagraphene [35,46] by the Stone-Wales transformation shown in Fig. S9 of Supplemental Material

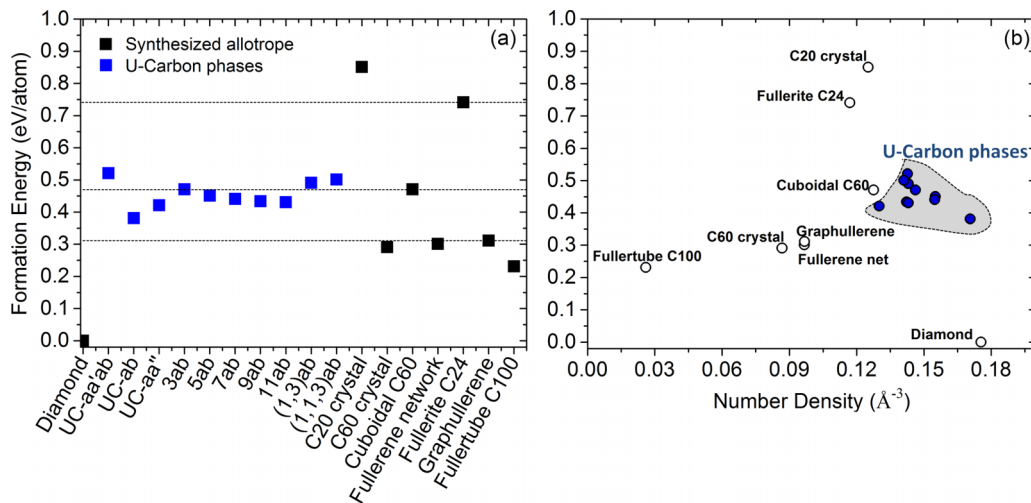


FIG. 5. Calculated formation energies (relative to diamond, eV/atom) of different carbon materials. (a) Formation energies the UC phases compared to a few synthesized carbon materials. (b) The formation energy vs. number density (number of carbons per unit volume) of the UC phases, in comparison with those of the experimentally observed carbon materials. The shaded area highlights the UC phases in the plot.

[19]. Both are buckled with two thirds of atoms being sp^2 and the other one-third being sp^3 bonded. Pentagraphene has a negative Poisson's ratio, while U-graphene has a positive one (0.056), as shown in Table S6 of Supplemental Material [19]. U-graphene has a band gap of about 2.0 eV while the composed UC is metallic. Pentagraphene has a significantly large band gap of 3.25 eV and its composed 3D materials, irrespective of stacking, are all semiconducting [46]. The UC structures exhibit high mechanical hardness and strength according to the calculated elastic constants, as shown in Table S5 of Supplemental Material [19]. The Young's moduli of all the structures are comparable to or higher than that of steel (200 GPa), tungsten (400 GPa) [47], and silicon carbide (450 GPa); especially the Young's modulus of the UC-ab structure is close to that of diamond (1050 GPa) [47]. The Poisson's ratios of the structures are in the range of 0.1–0.4, which are similar to those of concrete and glass. According to the Pugh criteria [48], UC-aa'ab and UC-ab are brittle like c-BN (~ 0.9) or diamond (~ 1.2) [49], while UC-aa'' is ductile like the silver metal. The UC structures, once experimentally realized, can

have many applications in energy materials, environmental treatment, catalysis, and medicine, with their exotic electronic, magnetic and mechanical properties originated from the special $sp^2 - sp^3$ hybrid topology by the various stacking of U-graphene layers.

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