Symmetry breaking and low energy conformational fluctuations in amorphous graphene

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Recently, the prospects for amorphous phases of graphene (α-g) have been explored computationally. Initial models were flat, and contained odd-member rings, while maintaining threefold coordination and sp² bonding. Upon relaxation, puckering occurs, and may be traced to the existence of pentagons, in analogy with the situation for fullerenes. In this work, we systematically explore the inherent structures with energy close to the flat starting structure. As expected, the planar symmetry can be broken in various ways, which we characterize for 800-atom model of α-g, always using local basis density functional techniques. The classical normal modes of various structural models are discussed, with an emphasis on imaginary modes indicating the evolution from flat to puckered. We also discuss very low energy conformational fluctuations akin to those seen previously in amorphous silicon, and reflect on the nature of the amorphous “ground state” within a network of fixed topology. For completeness, high energy modes were also computed, and are found to be associated with strained parts of the network.

1 Introduction A key feature of matter in a disordered state is the existence of large numbers of conformations with essentially degenerate energies, which may also be mutually accessible with small energy cost. This is in contrast with crystals, which possess long range order, few polymorphs and typically a deep energy minimum and large energy barrier. The ground state is thus sharply defined, and the only low energy excitations are phonons – transitions to other structures are prohibited. Where realistic models of disordered systems are concerned, few attempts have been made to quantitatively characterize the number, energetics, and proximity (in the sense of barrier) of these states. In his inherent structure formulation of statistical mechanics, Stillinger argued that the number of minima scales like \(N! \exp(\alpha N)\) [1], where \(N\) is the number of atoms in the model, and \(\alpha\) is a positive system-dependent constant. \(\alpha\) was estimated to be around 0.8 in a monatomic liquid [2], and flexible organic molecules exhibit larger \(\alpha\), as in the fragile glass former ortho-terphenyl where \(\alpha\) is around 13.14 [3]. For temperatures well below the melting point, these local energy minima are denoted inherent structures.

To further motivate this work, consider the following gedanken experiment. Consider a sequence of molecules with \(N\) atoms. It is well known that as \(N\) increases, the number of minima accessible to the molecule also increase, and such conformations are extensively studied in chemistry [4–7]. While it is unlikely that a rigorous theory quantifying these minima as a function of \(N\) can be formulated, it is clear from computer experiments that the number of minima grow drastically with \(N\). For most molecular systems it is difficult to be certain that the global minimum structure has been found in a simulation as there are so many metastable minima in which the system can be trapped. In this paper, we are concerned with the even more intractable problem of characterizing the minima, or potential energy surface of a disordered condensed matter system in both two-dimensional (2D) and three-dimensional (3D) cases. For a 3D crystal, if one introduces small random distortions and relaxes the disturbed system, it returns to exactly the same structure. This is not true for disordered systems. For 3D, we find there exits a continuum of metastable minima for α-Si, in which a number of tiny distortions of bond angles (and to a much lesser extent bond lengths) yield a distinct energy-degenerate conformation, which reveals the existence of an extraordinarily flat potential energy landscape (PEL). Fedders and Drabold showed that starting from a well-
relaxed α-Si:H model, quenching a set of “snapshots” during a constant-T MD simulation never returns to the exact initial state, instead they fall into minima that are topologically equivalent (e.g., with the same network connectivity), but with small variations in bond angles and bond lengths [8]. We find that the behavior in α-Si is consistent with α-Si:H. The case of α-g [9] is different in the following sense. Like α-Si, we find a continuum of essentially energy degenerate states in which minute (but “real”) variations in bond angles and bond lengths are displayed. Of course, such states retain identical connectivity since their energies are identical to within a few μeV. However, α-g also exhibits a variety of local energy minima associated with different puckering. These structures usually have similar energies (within ~10 meV), but a significant barrier separating them. Thus, the picture that emerges of the α-g energy landscape is a variety of inherent structures (with varying puckering) with slightly varying energies but substantial barriers between them, and in each of these basins (associated with a particular puckered state), an ambiguously defined minimum with small variations in bond angles and bond lengths accessible as we describe in detail below in Section 4.2. The structure of sp² amorphous carbon has been much discussed [10–12]. Our networks are quite different because of their strictly 2D nature, though this is not obvious in gross features such as the radial distribution function. Another class of potentially relevant structures are the “schwarzites”, 2D negative curvature versions of the graphene materials [13].

Beside the work described above on ultra-low energy excitations, we also discuss other phenomena peculiar to α-g. There has been intensive study in understanding the properties of crystalline graphene, but little is securely understood about amorphous phases. Recent electron bombardment experiments have revealed the existence of amorphous graphene [14, 15]. Clear images of regions of amorphous graphene have been published by Meyer [16]. In previous work, we observed that planar amorphous graphene is extremely sensitive to out-of-plane distortions [17]. Similar behavior has been verified in amorphous graphene by experiment and other calculations [18, 19]. We have found that very slightly different initial conditions (e.g., in the transverse coordinates) lead to very different puckering after relaxation [17]. These states exhibit little or no difference in topological properties, i.e., ring statistics and coordination number. However, the total energy difference between these metastable states is around 0.02 eV per atom, and the full width of the puckering along the transverse direction (between extrema) is around 6–7 Å.

A natural complement to these studies is an exploration of low-frequency classical normal modes. These modes turn out to be rather delocalized. As presented for two level systems, the tunneling between two equilibrium states triggers a number of low-energy excitations [20–22]. Details are discussed in Section 4.3. In Section 5, we summarize our findings about the degenerate states and localized imaginary-, low-, and high-frequency vibrational modes of amorphous graphene.

Figure 1 Comparison between crystalline and relaxed amorphous phases of graphene. Periodic boundary conditions are employed.
by very slightly breaking the planar symmetry (by randomly moving each atom by \(\sim 0.01 \text{ Å} \)) and again carrying out the minimization, the resulting structures were always puckered [17].

The amorphous Si model we employ is a realistic 64-atom model (64 \(\alpha\)-Si), generated by Barkema and Mousseau using a modified form of the WWW algorithm [25]. This model has ideal fourfold coordination and within the significant limitations of its small size, to our knowledge is not in significant contradiction with any experiment.

3 Procedure Our calculations are performed with the \textit{ab initio} program SIESTA [26], using pseudopotentials and the Perdew–Zunger parameterization of the local-density approximation (LDA) with a single-\(\zeta\) basis and Harris–Functional at a constant volume. Earlier simulations are described in Refs. [27] and [28]. To investigate the nature of minima on the potential energy surface, we employ a method proposed by Fedders and Drabold [8] similar to the conformational space annealing approach mentioned in Ref. [29], which has been used in locating and predicting low-energy conformations of various proteins [30–33]. First, starting with a perfectly relaxed model (in our case 800 \(\alpha\)-g, four MD simulations overcome tiny energy barriers and take a down-hill path to configurations with lower energy. Thus the flat 800 \(\alpha\)-g model can be considered as an exceptionally shallow basin on the PEL. The barrier between flat and puckered is a few \(\mu\text{eV}\) for this Hamiltonian.

4.2 Conformational fluctuations The quenching procedures at sequential timesteps yield basins on the PEL. Here we show the calculations at average temperatures of 500, 600, and 900 K. These results of MD runs at different temperatures exhibit consistency with each other. Figure 3 shows how these two autocorrelation functions vary with time. Since temperatures of all the MD simulations achieve equilibrium after 6.0 ps, here the autocorrelation functions are calculated with \(t_1 = 6.0\) ps. It appears \(\Delta \theta(t_1, t_2)\) and \(\Delta r(t_1, t_2)\) from three MD runs at different temperatures are qualitatively similar. They increase linearly and eventually fluctuate about a constant. The continuity of the curves in Fig. 3 suggests that there is a continuum of states, accessible albeit structurally varying only in very modest ways.

The total energy distributions of all the quenched supercells from MD runs at different temperatures are shown in Fig. 4. For temperatures other than 20 K, the total energy distributions exhibit several peaks. The minor peaks in Fig. 4 correspond with the annealing process of MD runs. The structure may be expected in these films [34]. As we have shown in Ref. [17], the original flat 800-atom \(\alpha\)-g model is exceedingly sensitive to transverse distortion, and then loses planar symmetry lowering the total energy of the supercell. In every case, even at \(T = 20\) K, the planar symmetry breaks and the system puckers: thermal disorder is sufficient to induce puckering. Figure 2 shows the relation between the total energy of the system and maximum separation of atoms along the normal direction (magnitude of puckering) in constant-\(T\) MD simulations at the indicated temperatures. In the language of PEL, starting from the flat 800 \(\alpha\)-g, four MD simulations overcome tiny energy barriers and take a down-hill path to configurations with lower energy. Thus the flat 800 \(\alpha\)-g model can be considered as an exceptionally shallow basin on the PEL. The barrier between flat and puckered is a few \(\mu\text{eV}\) for this Hamiltonian.

4 Discussion We break the discussion into three parts. First, the nature of pentagonal puckering, next, low-energy conformation fluctuations, and finally an analysis of the classical vibrational modes.

4.1 Symmetry breaking As conjectured by Cataldo et al. using purely topological analysis, fullerene-like structure may be expected in these films [34]. As we have shown in Ref. [17], the original flat 800-atom \(\alpha\)-g model is exceedingly sensitive to transverse distortion, and then loses planar symmetry lowering the total energy of the supercell. In every case, even at \(T = 20\) K, the planar symmetry breaks and the system puckers: thermal disorder is sufficient to induce puckering. Figure 2 shows the relation between the total energy of the system and maximum separation of atoms along the normal direction (magnitude of puckering) in constant-\(T\) MD simulations at the indicated temperatures. In the language of PEL, starting from the flat 800 \(\alpha\)-g, four MD simulations overcome tiny energy barriers and take a down-hill path to configurations with lower energy. Thus the flat 800 \(\alpha\)-g model can be considered as an exceptionally shallow basin on the PEL. The barrier between flat and puckered is a few \(\mu\text{eV}\) for this Hamiltonian.

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major peaks (labeled 3, 2, and 1 in Fig. 4) are derived from different puckering configurations sampled in the process of equilibration to constant $T$. Correspondingly as shown in Fig. 3, the fluctuations of autocorrelation functions (after thermal equilibrium is reached) reach an asymptotic state after of order 7.0 ps (1 ps from the initial equilibrated state). Each of the three peaks in Fig. 4 demonstrates a basin on the PEL of $\alpha$-g. These nearly degenerate quenched equilibrium states are trapped in distinct basins on the PEL, and quenched minima within one basin form a continuum metastable state around inherent structures. Details of variations in bond angles, bond lengths, and total energies of these metastable states are shown in Table 1. The total energy variation between the basins corresponding to the major three peaks are averaged as $1.405 \times 10^{-2}$ eV, which is one order of magnitude higher than the energy of fluctuations within a basin. In spite of their different energy scales, these quenched configurations belonged to distinct basins share identical local bonding. The only difference is that they pucker in distinct ways, as shown in Fig. 5.

Table 1 Average value and standard deviation of $E_{\text{tot}}/N_{\text{atom}}$, $\Delta r(t_1, t_2)$, and $\Delta \theta(t_1, t_2)$ of quenched configurations from MD runs in the time period from 3.6 to 8.0 ps, where $t_1 = 1.05$ ps.

| $T$ (K) | $E_{\text{tot}}/N_{\text{atom}}$ (eV) | $\sigma_{E_{\text{tot}}/N_{\text{atom}}}$ (eV) | $\Delta r(t_1, t_2)$ (Å) | $\sigma_{\Delta r}$ (Å) | $\Delta \theta(t_1, t_2)$ (°) | $\sigma_{\Delta \theta}$ (°) |
|---------|----------------------------------|----------------------------------|---------------------------|------------------------|---------------------------|------------------------|
| 500     | $-9.595 \times 10^{-2}$         | $8.855 \times 10^{-5}$          | $4.907 \times 10^{-2}$   | $2.551 \times 10^{-4}$ | $3.736$                   | $1.814 \times 10^{-2}$ |
| 600     | $-9.435 \times 10^{-2}$         | $9.917 \times 10^{-5}$          | $7.503 \times 10^{-2}$   | $1.973 \times 10^{-4}$ | $4.181$                   | $6.464 \times 10^{-2}$ |
| 900     | $-9.716 \times 10^{-2}$         | $2.091 \times 10^{-4}$          | $2.516 \times 10^{-2}$   | $8.08 \times 10^{-3}$  | $1.725$                   | $0.146$                 |

Figure 3 (online color at: www.pss-b.com) Time variation of two autocorrelation functions. This figure shows autocorrelation functions of $\Delta \theta(t_1, t_2)$ and $100\Delta r(t_1, t_2)$ for $t_1 = 6.0$ ps and $t_2$ varying from 6.0 to 7.95 ps. The temperatures are 500, 600, and 900 K. The functions appear to be continuous.

Figure 4 (online color at: www.pss-b.com) Total energy distribution functions of quenched supercells from MD runs under 20, 500, 600, and 900 K. The total energy of original flat $\alpha$-g is considered as 0 eV. Distinct structures correspond to different puckered states, broadening within each major peak from conformational variations. Three major peaks from MD runs at 600, 500, and 900 K are labeled as 1, 2, and 3 respectively.

Figure 5 (online color at: www.pss-b.com) Side view of two quenched configurations. Gray balls and sticks show the configuration from 900 K MD, and blue lines represent the one from 500 K.

Figure 6 (online color at: www.pss-b.com) Time variation of two autocorrelation functions for $\alpha$-Si. This figure shows autocorrelation functions of $\Delta \theta(t_1, t_2)$ and $100\Delta r(t_1, t_2)$ for $t_1 = 6.6$ ps and $t_2$ varying from 6.6 to 8.0 ps. The temperatures are 20, 300, and 500 K. The piecewise linear appearance is an artifact of coarse time sampling. The results are similar to Ref. [8].
For comparison, we repeat parallel calculations using 64 $\alpha$-Si model quenched from MD runs at 20, 300, and 500 K. The autocorrelation functions are shown in Fig. 6. The results are in agreement with Fedders and Drabold [8].

We see for $\alpha$-Si systems, there exists one general basin on the PEL (for a particular network connectivity), and the paths lowering the total energy on the PEL will eventually go into this basin, leading to inherent structures with minor changes in bond angles and lengths, and tiny energy scale.

Comparison between results of $\alpha$-g and $\alpha$-Si suggests that PEL of 3D system ($\alpha$-Si) is smooth and inherent structures are contained in one general basin. $\alpha$-g is similar within one puckered state.

For $\alpha$-g, an MD run at higher temperature (annealing) can overcome the energy barrier, and hop to a basin associated with a different puckered state. Also by investigating the correlation between topology and energy scale of these quenched supercells, it is revealed that lower total energy (stabler state) is associated with more puckered (higher surface roughness) configurations with small variation in bond lengths and angles from the original flat 800-atom model.

4.3 Classical normal modes

To investigate the vibrational modes in these supercells, we perform calculations of the dynamical matrix, its eigenvalues and eigenvectors, for the original flat 800 $\alpha$-g model, and two quenched configurations with certain regions puckering in opposite directions, designated “pucker-up” and “pucker-down” models, as shown in Fig. 7. The dynamical matrix was constructed from finite difference calculations (using six orthogonal displacements of 0.04 Bohr for each atom). We also perform the phonon calculation for an 800-atom crystalline graphene model (for related calculations on very large fullerenes, see Ref. [36]). The vibrational density of states (VDOS) of 800 crystalline model, pucker-up and -down 800 $\alpha$-g models are shown in Fig. 8. The VDOS result of crystalline graphene shows good agreement with a published calculation [35]. In Fig. 8 at a frequency near $1375 \text{ cm}^{-1}$, the spectrum of crystalline graphene has a minimum. In contrast the spectrum of two puckered supercells achieve a local maximum. Thus Raman scattering experiments are expected to provide a way to distinguish crystalline and amorphous graphene. There is no difference in the spectrum between pucker-up and -down 800 $\alpha$-g models. In a mixed sample containing 3D amorphous carbon, this discernible difference from ordered graphene near $1375 \text{ cm}^{-1}$ is probably unhelpful because of the many modes seen in various phases of $\alpha$-C (with varying sp$^2$/sp$^3$ ratio) near the relevant energy [37–39].

In the original flat 800-atom $\alpha$-g model, the eigenvectors with imaginary eigenvalues have large components along the normal direction of the graphene plane (at least four orders of magnitude higher than longitudinal components). These imaginary-frequency modes are localized on pentagons in the network: two example are shown in Fig. 9. As shown in Fig. 9c and d, these imaginary-frequency modes are localized near structures that lead to puckering. As shown in Ref. [17], it is pentagons that lead to puckering and symmetry breaking. Thus these imaginary-frequency modes are an indicator of the instability of the flat 800 $\alpha$-g model.

In the puckered models, we observe modes with a low frequency, around 14–20 cm$^{-1}$, reminiscent of “floppy modes” proposed by Phillips and Thorpe [40, 41]. The structures of these low-frequency modes are quite complex, as shown in Fig. 11. These modes are rather extended, and have significant weight on pentagonal puckered regions and large rings, analogous to what Fedders and Drabold have seen in $\alpha$-Si:H [8]. The observed energy scale of these low-frequency modes is around a few meV, almost half of the lowest frequency of the acoustic phonon modes in a crystalline graphene with same size. As proposed in the theory of “two-level systems”, there exists a distribution of low-energy excitations, caused by tunneling of atoms between nearly degenerate equilibrium states [21, 22]. Goldstein pointed out that the dynamics could be separated into two categories: vibrational motion about a minimum on PEL and transitions between minima [42]. Then these low-frequency modes might be triggered by transitions between degenerate minima within one basin on the PEL. As shown in Section 4.2, the energy variations between minima within one basin is in the order of $10^{-5}$ eV, and the energy difference between basins is in the order of $10^{-7}$ eV. The energy scales of these low-frequency modes (~meV) are
sufficient to drive conformational fluctuations, but not high enough to overcome the energy barrier between different basins (quenched states) on the PEL.

In the high-frequency domain, there also exist highly localized high-frequency modes in the puckered configurations. These modes are triggered by the pentagonal defects and are highly localized, as shown in Fig. 12. This result is consistent with Biswas et al. [43] and Fedders et al. [44], who have shown strain and topological defects are active at highest frequencies.

We also compute the specific heat $C(T)$ using VDOS information [45]:

$$C(T) = 3R \int_0^{E_{\text{max}}} \left( \frac{E}{k_B T} \right)^2 \frac{e^{E/k_B T}}{(e^{E/k_B T} - 1)^2} g(E) dE, \quad (3)$$

where $g(E)$ is normalized VDOS. For room temperature (300 K), specific heat of flat, pucker-up and -down 800 $\alpha$-g models are 25.151, 18.879, and 18.702 J K$^{-1}$ mol$^{-1}$, respectively. The temperature dependence of $C(T)$ is shown in Fig. 10. This is presumably an academic result as it is currently hard to imagine an experiment for $C(T)$ for this 2D system.

5 Conclusions In conclusion, we have found that $\alpha$-g has a rich and interesting energy landscape. We observe distinct energy scale of basins ($\sim$10 meV) associated with different puckered configurations and then within such a configuration, an ambiguous energy minimum with a continuum of bond angles and bond lengths with energy
scale (\textsim\ few \mu eV) and a nearly flat PEL. Within a given puckered configuration, this continuum is much like what was seen for \alpha-Si:H in 1996 [8].

Vibrational calculations reveal the existence of localized imaginary-frequency modes in flat 800 \alpha-g model. These modes are localized on pentagons and play the key role in losing planar symmetry and forming pentagonal puckering structures. We find delocalized low-frequency phonon modes, similar to floppy modes, which have substantial weight on defects and share the same energy scale as the energy difference between adjacent basins on the PEL. Thus these low-frequency modes are triggered by the transition between adjacent energy minima. Some high-frequency modes are detected and highly localized on puckered regions and large rings.

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