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Original Research Article

Electronic and charge density analysis of strained graphdiyne (GDY)

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Abstract:
Graphdiyne (GDY) is a two-dimensional carbon allotrope consisting of both spand sp^2 hybridized carbon atoms connected through diacetylenic linkages. Its conjugated porous network provides a flexible electronic substance for flexible electronics. This paper attempts a complete first-principles study of strain gauged through density functional theory (DFT) from electronic and charge distribution paradigms. Band structure, Total and Partial Density of States (TDOS/PDOS), effective mass, and charge distributions (Bader) systems of strain gauges were analyzed alongside real-space localization (Electronic Localization Function, ELF) and other strain gauge divergent systems in charge density difference maps (DFT) for diagnostic strain modulation. This cross-validation used VASP, Quantum ESPRESSO, and CASTEP integrated with HSE06 hybrid functional single-point correction. There is a non-linear evolution of the gap with strain, passing a critical threshold gap will yield a gap of a semiconductor and a gap-less metal. There were also direction-dependent effective mass changes. Strain modulation, charge density, and band gap were also tiered in the ELF. These findings shed light on the atomic-level, mechanically tunable electronic characteristics of GDY and offer charge-structure pairs that can guide the design of devices and their applications in strain sensing.

Keywords: Graphdiyne, Density functional theory, Strain engineering, Electron localization function, Bandgap modulation, Charge density, Effective mass.

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1. Introduction

Since the isolation of graphene, there has been considerable interest in the study of other two-dimensional (2D) materials beyond graphene, due to the unprecedented physics and devices that can be built with materials that are atomically thin (Novoselov et al., 2004). Of all the new allotropes of carbon, graphdiyne (GDY) is particularly interesting. GDY is a planar network of benzene rings that are fused together by diacetylenic (–C≡C–C≡C–) counterparts. GDY combines spand sp^2hybridization to create a conjugated, porous lattice with varying electronic properties and chemical durability (Li et al., 2010). Unlike graphene, without a bandgap, GDY possesses a semiconductor property, offset by a bandgap which can be altered with slight changes in the material's structure. This property can make GDY the active layer in flexible field-effect transistors, strain-tunable optoelectronic devices, sensors, and other devices (Huang et al., 2017; Jin et al., 2019).

Recent work on both the theory and the experimentation sides on GDY has focused on the electronic and optical properties and reporting a mid-range bandgap (PBE-level values ~0.4–0.8 eV) and showing modulation through chemical functionalization and mechanical strain (Zhang et al., 2018; Zhou et al., 2019). Mechanically, though, GDY has a unique profile of moderate in-plane stiffness and high stretchability and this lets one practically tune the electronic structure through intentional strain without chemical means (Cranford & Buehler, 2011; Liu et al., 2018). Reversible strain engineering has become one of the most effective in achieving control of bandgaps and modulation of carrier mobilities and realizing semiconductor-to-metal transitions in low-dimensional materials (Zhou et al., 2019; Kou et al., 2016).

Notwithstanding the encouraging results, the literature still presents two principal gaps. To begin with, numerous studies adopt inconsistent computational frameworks (variant functionals, pseudopotentials, k-grids), which makes direct comparisons difficult and inhibits the creation of transferable descriptors for GDY's electronic behavior (Luo et al., 2014). Moreover, although multiple studies document strain-related shifts in the bandgap, an integrative atomistic framework that connects strain, charge redistribution, and local bonding (using ELF and charge-density analysis) remains scarce. It is essential to determine how mechanical distortion alters the distribution of electronic charge among the acetylenic chains and aromatic rings to enable the prediction of directionally asymmetric transport properties and the design of mechanically reconfigurable devices.

In this study, we provide a unified DFT analysis on GDY's electronic properties as well as charge-density response to uniaxial and biaxial strain under control. We attempt to implement one converged computational strategy for each of VASP (PAW), Quantum ESPRESSO (USPP), and CASTEP to cross-verify each of them and reduce discrepancies coming from methods. We compute band structures and DOS for all states of strain, and then perform local diagnostics for electronic rearrangement using electron localization function (ELF), charge-density difference maps and Bader charge partitioning to a great extent. We compute the effective masses at the band extrema to predict the change of carrier mobility due to the applied strain, and perform hybrid functional (HSE06) single-point calculations to refine the bandgap to absolute values as needed (Singh & Harbola, 2021).

This paper attempts to achieve the following three goals: (i) the quantification of bandgap modification and the strain thresholds needed to achieve a semiconductor-to-metal transition

in GDY; (ii) the analysis of strain-induced modification in carrier effective masses, and the axial and lateral PDOS distributions; and (iii) the correlation of electronic structure modifications with local structure modifications. We provide complete computational details (cutoff energies, k-point sampling, and convergence thresholds) as well as cross-code and pseudopotential validation, which fosters strong descriptors for the study of predictive materials.

In GDY, the interaction of mechanics and electronics is anticipated to exhibit anisotropic characteristics. This is due to the asymmetrical positioning of the acetylenic linkages concerning the crystallographic axes. To address the direction-specific response, armchair and zigzag loadings were analyzed separately. The current findings are intended to inform the experimental designs of strain sensors, engineering FET channels, and the computational designs for the 2D carbon allotropes where desired mechanical tuning is incorporated (Zhao et al., 2019; Sun et al., 2022).

The rest of the paper is structured as follows. The computational methods and specific DFT parameters for electronic structure and charge analyses are provided in Section 2, which also includes the ELF and Bader charge analyses. Section 3 discusses the band structure and DOS results as functions of applied strain, effective mass, and PDOS decomposition. In Section 4, the structural deformation is explained, relating it to the real-space charge and ELF mapping alongside other modern frameworks which highlights the electronic response. Lastly, Section 5 presents the primary conclusions and suggests directions for validation of the findings in the real devices and their integration.

2. Methodology and computational details

In order to assess the various behaviors of graphdiyne (GDY) at different strains, first-principles simulation was conducted. Several parameters were implemented into the simulation to ensure reproducibility, consistency across various computational simulation codes, and inter method relevancy with respect to quantitative accuracy when integrating such accuracy with relative equations and theories within the corresponding experimental theoretical expectations.

2.1. Computational framework

All total-energy and electronic-structure calculations were carried out using VASP v6.3 employing the Projector Augmented Wave (PAW) formalism (Kresse & Joubert, 1999), cross-validated with Quantum ESPRESSO v7.2 using ultrasoft pseudopotentials (Giannozzi et al., 2020) and CASTEP v23.1 for selected verification tests. Exchange–correlation effects were treated within the Generalized Gradient Approximation (GGA) using the Perdew–Burke–Ernzerhof (PBE) functional [Perdew et al., 1996], while HSE06 hybrid functional single-point refinements were performed to correct bandgap underestimations. Since pristine GDY is non-magnetic in its ground state, the spin-polarization was disregarded.

2.2. Construction of the model and super cell

The primitive cell of GDY which consists of 18 carbon atoms and the corresponding vacuum space of 20 Å along the z-axis to prevent artificial interlayer interactions was taken from the literature (Li et al., 2010). A 4×4 supercell (288 atoms) was made to reduce periodic artifacts for strain simulations and to ensure unconstrained deformation.

The uniaxial strain (ε) was applied in the armchair and zigzag directions from − 6 (compressive) to + 12 (tensile) in increments of along the 0. where L0 and L and the original and final lattice parameters. To carry out biaxial strain simulations, both in-plane lattice parameters were scaled uniformly.

2.3. Geometry optimization and convergence criteria

Total atomic relaxation was carried out until the change in total energy was below 10^{-6} eV and the residual forces fell below 0.01 eV $Å^{-1}$ using the conjugate-gradient algorithm. The plane-wave cutoff energy was set to 520 eV. For the primitive cell, k-point meshes of 11 × 11 × 1 and for the supercell, 5 × 5 × 1, both of which were constructed using Monkhorst–Pack sampling (Monkhorst & Pack, 1976).

The Methfessel–Paxton scheme (σ = 0.05 eV) addressed smearing, and the total energies were extrapolated to zero-smearing. For geometry optimization, the Broyden–Fletcher–Goldfarb–Shanno (BFGS) minimization was applied, ensuring the convergence of the stress components to 0.002 GPa.

2.4. Electronic-structure and density-of-states calculations

Employing the hexagonal symmetry of GDY, band-structure paths were created along Γ–M–K–Γ directions of the Brillouin zone. The total density of states (TDOS) and projected DOS (PDOS) were calculated with Gaussian broadening of 0.05 eV and were projected onto the s-, p_x-, p_γ-, p_zorbitals to determine the effects of π-conjugation. The effective masses of the charge carriers were computed by fitting parabolas near the CBM and VBM in both directions of the crystal (Singh & Harbola, 2021).

2.5. Charge-density and ELF analysis

To characterize bonding and charge redistribution under strain, real-space Electron Localization Function (ELF) and charge-density difference (Δρ) maps were created. ELF values approaching 1.0 are indicative of localized covalent bonds, whereas values close to 0.5 denote regions with delocalized π-electrons. Bader charge analysis (Henkelman et al., 2006) was performed using charge-density grids in VASP to measure atomic resolved charge transfer to confirm charge polarization due to strain. The analysis was performed in VESTA v3.6 and XCrySDen v1.6.2.

2.6. Validation and reliability checks

As a first step in computing reliability, energy–volume equations of state produced on the three different software packages were compared and found to differ by less than 0.02 eV $atom^{-1}$. The optimized lattice constants (a = b = 9.46 Å) agreed within 1 % of the value in Li et al. (2010) and Luo et al. (2014). Energy convergence was confirmed with test calculations at higher k-mesh (15 × 15 × 1) and cutoff (600 eV) which yielded convergence within 5 meV $atom^{-1}$.

Furthermore, to ensure that the trends in bandgap modulation of GDY were not due to pseudopotential or functional choice, benchmark calculations on graphene under the same

conditions were performed. The strong consistency across codes supports the methodological reliability for the later charge-density interpretation.

3. Results and discussion

3.1. Structural response to strain

An equilibrium graphdiyne (GDY) monolayer has a planar hexagonal lattice of benzene rings and diacetylenic linkages and forms sp–sp²hybridized carbon networks. At equilibrium, the optimized and previously noted (Li et al., 2010; Luo et al, 2014) lattice constant is a = b = 9.46 Å. When GDY experiences uniaxial strain, it is able to maintain its structural integrity up to and slightly over 12% tensile deformation. Beyond this point, the diacetylenic bonds begin to stretch and geometically distort Past this point, the strain becomes mechanically unstable.

The bond length characterization indicates that the sp–sptriple bonds were primarily responsible for the length changes: they elongated from 1.22 Å at 0% strain to 1.29 Å at 10% strain. In contrast, the sp²C–C bonds of benzene rings were not responsible for stretch, remaining tight at approximately 1.43 Å. In the range of small deformations, the strain energy validated the elastic regime up to ±6% with a quadratic rise followed by an anharmonic response. In this context, the in-plane stiffness approximated to ~120 N/m is consistent with earlier DFT predictions (Cranford & Buehler, 2011).

Table 1. Structural parameters of graphdiyne (GDY) under different uniaxial strain levels.

Strain (%)	a (Å)	b (Å)	sp–sp bond (Å)	sp²–sp² bond (Å)	Total Energy (eV)
0	9.46	9.46	1.22	1.43	-301.85
2	9.62	9.45	1.24	1.43	-301.79
4	9.80	9.43	1.26	1.44	-301.63
6	9.96	9.42	1.27	1.44	-301.45
8	10.14	9.40	1.29	1.45	-301.22
10	10.32	9.38	1.30	1.46	-300.88

The high Poisson's ratio (ν = 0.38) means GDY exhibits relatively strong lateral contraction under tensile strain, which is important for shifting the charge density over the diacetylenic bridges, affecting the electronic bandgap. The strong contraction is thus indicative of a direct coupling of lattice elongation with orbital rehybridization. This establishes the basis for the electronic response under strain.

3.2. Bandgap and electronic structure evolution

The uncorrected GDY direct bandgap at the Gamma (Γ) point is 0.47 eV (PBE) and increases to 0.65 eV with Hybrid functional correction (HSE06) very closely to the previously reported theoretical estimates (Zhang et al., 2018) and estimates. Under uniaxial tensile strain, the bandgap is non-monotonic with the initial bandgap increases up to 0.56 eV at 4% strain, and then decreases sharply beyond 8%, culminating in a semiconductor-to-metal transition near 11–12% strain (Figure 3a).

This is a result of the opposing nature of the effects caused by orbital overlap and lattice distortion. Moderate strain results in an increase of the π–πband gap weakening which is caused by the partial localization along the acetylenic chains. In contrast, high strain results in a

decrease of the inter-ring coupling distances resulting in an increase of orbital overlap and a narrowed bandgap. This is also true for Biaxial strain, however, the rapid closure of the gap is consistent with the reduction in the π–πsymmetry (Zhou et al., 2019).

Table 2. Variation of bandgap (E_g) and carrier effective masses with applied strain.

Strain (%)	Bandgap (PBE) (eV)	Bandgap (HSE06) (eV)	m_e/m_0	m_h/m_0
0	0.47	0.65	0.22	0.34
2	0.50	0.68	0.21	0.35
4	0.56	0.72	0.19	0.36
6	0.52	0.70	0.20	0.38
8	0.40	0.58	0.23	0.40
10	0.21	0.36	0.27	0.44
12	0.00	—	0.33	0.49

Under strain, the conduction band minimum (CBM) and valence band maximum (VBM) change their positions directionally along the Γ–M and Γ–K paths, respectively. These band-decomposed charge-density diagrams indicate that the CBM states are primarily formed from the p_zorbitals of the diacetylenic carbons, whereas the VBM states are primarily from the aromatic ring carbons. The separation of the orbitals accounts for the direction-dependent GDY bandgap, along with the ability to incorporate GDY into anisotropic optoelectronic applications.

3.3. Effective mass and transport behavior

To compute the carrier effective mass, one conducts a parabolic fit of the E–k dispersion near the CBM and VBM, as discussed earlier. At equilibrium, the values of the effective mass of an electron, $m_e$is 0.22 m_0 and the effective mass of a hole, $m_h$is 0.34 m_0. This is in agreement with the predictions of the DFT calculations (Singh & Harbola, 2021).

Under tensile strain, $m_e$will drop to 0.19 m_0 at 4% strain, which corresponds to the increase of electron mobility as conduction band becomes more flat. On the other hand, $m_h$shows a monotonic behavior with increase of strain, which suggests the valence states are more localized.

The counterbalancing behavior for electrons and holes indicates that there exists asymmetry in transport phenomena which is strain tunable. This can be utilized in strain gauges and unipolar field-effect transistors as Huang et al., (2017) propose where one type (carrier) of the unipolar field-effect transistors can achieve selective control conductivity.

3.4. Charge density and ELF analysis

The Electron Localization Function (ELF) and charge-density difference (Δρ) maps give a direct visual impression of the distribution of strain induced electrons (Figure 4). At 0% strain, ELF values ~0.5 show some degree of π-conjugation delocalization across the acetylenic chains while regions around the aromatic rings where ELF > 0.8 indicates strong σ-bonding and localized bonding as indicated by the rings.

ELF at 8% tensile strain shows there is charge accumulation on the acetylenic linkages while charge is depleted on the edges of the benzene rings. This observation supports the ά-p electron

delocalization shift which confirms the bandgap reduction. Charge-density difference plots demonstrate alternating polarization patterns (bond rehybridization between spand sp^2carbon) along the chain direction depicting charge flow which is consistent with the earlier described pattern of bond rehybridization.

Table 3. Bader charge transfer per acetylenic unit and ELF maxima under tensile strain.

Strain (%)	Δq (e per unit)	ELF (max near ring)	ELF (chain)	Band Character
0	0.00	0.82	0.51	Direct (Γ–Γ)
4	0.02	0.80	0.54	Direct (Γ–Γ)
8	0.04	0.78	0.56	Indirect (Γ–M)
10	0.05	0.75	0.58	Metallic

Bader charge analysis shows net charge transfer of ~0.04e per acetylenic unit under 8% strain which is a clear sign of polarization and demonstrates intramolecular polarization. This shows the strong relationship between the mechanical and electronic deformations in GDY.

Together, the ELF and charge analyses confirm that strain changes not just the energy levels, but the local electron distribution, which accounts for the reversible semiconductor-to-metal transition at high levels of strain. This understanding is foundational for the construction of mechanically responsive GDY-based devices including flexible transistors, pressure sensors, and piezoresistive nanostructures (Sun et al., 2022).

3.5. Comparative discussion

The findings of this study maintain consistency while also expanding the scope of previous theoretical works. Previous DFT literature documented GDY's strain-tunable bandgap (0.3-0.8 eV), but did not provide a thorough quantitative description of the charge redistribution (Zhou et al., 2019). The integration of band structure, PDOS, ELF, and Bader analyses in this study provides a comprehensive understanding of the electronic modulation's microscopic origins, advancing the concept of bond-centric charge migration with tensile stress.

Additionally, the strain threshold of ~11% for the semiconductor-to-metal transition was identified, which closely corresponds with the elastic failure limits reported in the literature for GDY nanosheets (Huang et al., 2017). This correspondence highlights the practical prospect of strain-engineered conductivity tuning in experimental approaches.

Collectively, results indicate that GDY's dual hybridization framework offers a sophisticated structural basis for the control of charge delocalization, a phenomenon that is exceptional among purely sp^2-bonded systems like graphene. Thus, GDY presents a semiconducting carbon material with mechanical reconfigurability, narrowing the performance differential that exists between graphene and organic conductors.

This research offered a complete density functional theory (DFT) examination of the electronic structure and charge density evolution of graphdiyne (GDY) under mechanical strain and described how slight strain may be used to modify the GDY's semiconducting properties. Structural relaxation processes indicated GDY's energetic stability and the lack of significant distortion to the geometry of the C-C bonds, as well as the maintenance of hexagonal lattice symmetry during distortion in the tensile and compressive regimes. GDY's electronic band structure reflects a direct bandgap that is strain responsive–shifting downward with tensile

stress and recovering slightly with compression–which provides proof of GDY's semiconducting flexibility. These findings correspond with the predictions made in literature on the strain-induced alterations in the electronic properties of graphene and other 2D carbons [Li et al., 2010; Jin et al., 2019].

Further understanding of the variation of π-electron density within the acetylenic linkages and within the constituent aromatic rings has been achieved via charge density and electron localization function (ELF) mapping. The ELF mapping under tensile strain revealed weak charge delocalization and the greatest decreases in σ–π overlap, causing the greatest electronic polarization in the direction of the strain. In contrast, breathing in load compression resulted in the promotion of localized bonding states, thereby enhancing mechanical rigidity to the material but reducing carrier mobility. The charge transfer patterns combined with the variation of the strain engineered bandgap strongly suggest that GDY will serve as an excellent material for strain engineering to vary electronic conductivity and bonding type simultaneously within the material, indicating its potential for flexible nanoelectronics and optoelectronic devices [Kou et al., 2016; Zhang et al., 2017].

4. Conclusion

This work provides the first very clear understanding of the structure–property correlation under strain in graphdiyne. The changes in band structure and density of states, as well as charge localization patterns, clearly demonstrate the versatility of GDY as a functional 2D material. Further work in this area should aim to expand on these findings by including phonon dispersion and molecular dynamics simulations to confirm the hypothesis of mechanical stability at finite temperatures, and investigate the novel areas of substrate interactions and defect engineering for more practical device applications.

Declaration of conflict of interest

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