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Magnetic localized states and tunable magnetism of single vacancies in generalized chiral graphene nanoribbons

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Abstract

In this paper, we obtain the generalized chiral edges of graphene nanoribbons, through longitudinal unzipping of carbon nanotubes. After analysing the stability and magnetic localized states of the generalized chiral edges based on first-principles calculations, we find the novel phenomena will arise, i.e., antiferromagnetic order in one edge and ferromagnetic order between different edges. And furthermore, the vacancy in the bulk can induce or enhance the magnetic states in the edges.

1. Introduction

Graphene, a single-layer two-dimensional carbon material, has attracted enormous interest due to its novel electronic properties and potential device applications [1–3]. Graphene nanoribbons (GNRs), cut from the graphene, are quasi-one-dimensional materials whose properties are dominated by the geometry of their edges [4, 5]. There are two well-known types of edges for GNRs, namely, zigzag and armchair edges. Different from the armchair GNRs (AGNRs), the zigzag GNRs (ZGNRs) have localized edge states at the Fermi level [6, 7]. Besides the basic zigzag edges and armchair edges, there are also several specific edge structures have been studied, such as Klein edges [8–11], Stone-Wales edges [11, 14–18] and chiral edges [19–28].

The major approaches for synthesis of GNRs include cutting graphene by lithographic techniques [29–31], synthesis from polycyclic molecules [32, 33], etching down of graphene [34, 35] and unzipping of carbon nanotubes (CNTs) [36–43]. Compared with the former three methods, unzipping CNTs offers a better solution to obtain GNRs with relative smoother edges. However, the unzipping path cannot follow one zigzag or armchair high-symmetry orientation, leading to the edges of GNRs composed of several zigzag and armchair chains. Major previous theoretical works usually concentrate on the ideal chiral edges, which is composed of one zigzag chain and one armchair chain. However, due to the effects of adatoms, inevitable impurities, substrates, etc., even the ideal chiral edges are difficult to obtain experimentally. As a result, if we want to study the properties of GNRs unzipped from GNTs, we must consider the generalized chiral edges which contain several zigzag and armchair chains with different orientations.

In this paper, we analyse the unzipping rules of single-walled CNTs (SWCNTs), and obtain the generalized chiral GNRs (GCGNRs) which possess several zigzag and armchair chains. Since the ZGNRs are well-known for their magnetic edge states, we should ask the question: Whether edge states can survive or not in the GCGNRs? Interestingly, we find that near the junctions of neighbouring zigzag chains with different orientations, the antiferromagnetic order in one edge and the ferromagnetic order between different edges can emerge. This is totally different from the well-known edge states of ZGNRs in which the ferromagnetic order can only exit in one edge and the antiferromagnetic order can only exit between different edges. Furthermore, we study the single vacancy effects in the GCGNRs, and find that the emergent magnetic properties can be explained by the famous Lieb’s theorem [44]. The more interesting thing is that the magnetism of bulk vacancies may induce or
enhance the edge states of the GCGNRs. For instance, if the zigzag chains in the edge are too short to support edge modes, there will be little edge states in the ideal GCGNRs. However, when the single vacancy defect is introduced near the edge, depending on the sublattice type of the vacancy and edge atoms, the localized states near the vacancy may induce or enhance the edge states due to the interactions between them.

2. Calculation methods

Our spin-polarized calculations are based on the density function theory (DFT) and are implemented in the CASTEP code [45]. The ultrasoft pseudopotentials [46], Perdew–Burke–Ernzerhof (PBE) exchange-correlation functionals and generalized gradient approximation (GGA) are adopted [47]. In our calculations, the plane-wave energy cutoff is taken to be 750 eV, and a Monkhorst–Pack [48] $3 \times 1 \times 1$ $k$-points mesh is adopted. Convergence with respect to the plane-wave cutoff energy and $k$-point sampling has been carefully checked. In both the energy band and density-of-states (DOS) calculations, the $3 \times 1 \times 1$ $k$-points mesh are adopted. It should be pointed out that in the DOS calculations, we have increased the $k$-points mesh to, which leads to negligible energy differences, implying is enough for high accuracy.

The carbon atoms at the edges of the GCGNRs or near the single vacancy defects have been passivated by hydrogen atoms, and the primitive cell contains 108 carbon atoms (or 107 carbon atoms if single vacancy exists) and dozens of hydrogen atoms. The horizontal and vertical distances between neighboring GCGNRs are set to be 15 and 12 Å, respectively, which leads to negligible interactions between them. All structures are full relaxed. During the geometry optimization, the convergence threshold for the maximum energy change, the maximum force, the maximum stress, and the maximum displacement are $1 \times 10^{-3}$ eV/atom, 0.03 eV/Å, 0.05 GPa, and 0.001 Å, respectively. When we try to figure out which kind of generalized chiral edges are more stable, we design four typical GCGNRs with the same numbers of carbon and hydrogen atoms, thus we can directly compare the total energies of each structure.

3. Results and discussion

3.1. Unzipping rule of SWNTs to GCGNRs

The structure of a SWCNT can be defined by the chiral vector which is the linear combination of the two lattice vectors of graphene $C_0 = n_1a_1 + m_1a_2$. $C_0$ is corresponding to the section of the nanotube perpendicular to the nanotube axis, and determines the diameter and chiral angle $\theta$ of the nanotube where $\theta$ is defined by $\cos \theta = C_h \cdot a_1/|C_0||a_1|$. Generally, a graphene sheet can be rolled into a tube in three distinct ways: (1) armchair graphene nanotube (AGNT): when $n_1 = m_1, C_0 = (n_1, n_1)$ and $\theta = 30^\circ$; (2) zigzag graphene nanotube (ZGNT): when $m_1 = 0, C_0 = (n_1, 0)$ and $\theta = 0^\circ$; (3) chiral graphene nanotube (CGNT): $C_0 = (n_1, m_1)$ and $0^\circ < |\theta| < 30^\circ$.

Both ANTs and ZNTs are called achiral nanotubes (ACNTs). For simplicity, we theoretically discuss the unzipping rules of ACNTs to GCGNRs in this paper, and study the magnetic states of the GCGNRs after the unzipping.

For a clear demonstration of unzipping processes, we plot the AGNT and ZGNT in figures 1(a) and (b) respectively, where the left and right edges of each should be rolled and connected with each other. Rather, the longitudinal direction of the GNT is along the $y$ direction. Any longitudinal unzipping process of the GNT can be simulated by a polygonal line connecting the upper and lower boundaries. The polygonal line, linked by several straight lines, describes the unzipping path. Due to the specific structure of the graphene, the intersection angle between the straight lines and $y$ axis $\phi$, can only be $0^\circ$, $30^\circ$, $60^\circ$, $90^\circ$, $120^\circ$, $150^\circ$ and $180^\circ$. Unzipping along the lines with different $\phi$ led to GNRs with different edges, which is summarized in table 1. We can see from table 1 that: (1) for both the AGNT and ZGNT, when $\phi$ is changed by $30^\circ$, the type of corresponding edge after the unzipping should be changed; (2) for the same $\phi$, the type of corresponding edges after the unzipping are different for the AGNT and ZGNT.

Because any polygonal line connecting the upper and lower boundaries simulates one whole unzipping path, the GNRs unzipped from the GNTs can have very complex edges. Obviously, these complex edges are far beyond the conventional chiral edges [19–28] which only contain one zigzag chain and one armchair chain in each unit cell. As a comparison, our edges contain many zigzag and armchair chains with various orientations. In this sense, our edge can be viewed as a generalized chiral one, and we call the obtained GNRs GCGNRs. It should be pointed out that, since the GCGNRs contain many different zigzag and armchair chains, it’s difficult to define the unified chiral vector [19, 21] for GCGNRs.

In practice, many variations of generalized chiral structures can exist in the edges. To figure out which kind of generalized chiral edges are more stable, we design four typical GCGNRs with different degrees of bending in figure 2. To facilitate the calculations, we suppose the GCGNRs are periodic. All these GCGNRs have been passivated by hydrogen atoms and possess the same number of atoms in one unit cell. The carbon atoms marked
red (blue) belong to the armchair (zigzag) chains. We compare total energies of these GCGNRs in table 2 and find that the GCGNRs with smoother edges have lower energies. As a result, the bending edges are not energetic favorable in ideal unzipping processes. This phenomenon can be explained as follows: formations of sharply curved junctions between different chains will cost more energies, thus the flat edges would be energetic favorable. However, it must be pointed out that, due to the effects of inevitable adatoms, impurities, etc., the unzipping paths may be changed for lowering energies. Consequently, even though the GCGNR in figure 2(a) has the lowest energy, the other three GCGNRs in figure 2(b)-(d) may also appear.

3.2. Magnetic edge states of ideal GCGNRs

Since the edges of GCGNRs are very complex, we choose one typical case in figure 3, which is periodic in the y direction. For AGNTs, successive $\phi = 0^\circ$ and $\phi = 60^\circ$ unzipping paths will led to such kind of generalized chiral edges. The main feature of this edge is that two zigzag chains with different $\phi$ cross with each other. In figure 3(a), zigzag (armchair) chain n is abbreviated to Zig-C-n (Arm-C-n). The generalized chiral edges which are composed of two neighbouring zigzag chains, i.e., zigzag chain 1 and 2 (3 and 4) in the upper (lower) edge. To facilitate the calculations, we choose the periodic boundary condition, thus the zigzag chain 1a and 1b (3a and 3b) belong to the same zigzag chain 1 (3). As a result, two novel phenomenons emerge: (1) antiferromagnetic

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**Figure 1.** (a) and (b) describe the unzipping paths of the AGNTs and ZGNTs, respectively. Any polygonal line connecting the upper and lower edges simulating a longitudinal unzipping process of the GNTs.

**Table 1.** Unzipping rules of AGNTs and ZGNTs

| $\phi$   | AGNT  | ZGNT  |
|---------|-------|-------|
| 0°      | Zigzag| Armchair |
| 30°     | Armchair| Zigzag |
| 60°     | Zigzag| Armchair |
| 90°     | Armchair| Zigzag |
| 120°    | Zigzag| Armchair |
| 150°    | Armchair| Zigzag |
| 180°    | Zigzag| Armchair |
coulping can emerge in one edge. In figure 3(a), spin-up (red) and spin-down electrons (blue) localize in the GCGNR, showing the antiferromagnetic coupling between the zigzag chain 1 and 2 (3 and 4) in the upper (lower) edge. This is because the edges of the two neighbouring zigzag chains belong to the different sublattices. However, for conventional ZGNRs, only ferromagnetic order exits in each edge. (2) Ferromagnetic order emerges between the two edges near the junction, i.e., ferromagnetic coupling between the zigzag chain 1 and 4 (2 and 3), which is attribute to the fact that the atoms at the two different edges belong to the same sublattice. This is also different from the ZGNRs where only antiferromagnetic order exits between the two edges. To the best of our knowledge, these two novel features of magnetic edge states of GNRs have never been reported before. Figure 3(b) shows the band structure and density of states (DOS) of the GCGNR. The blue solid lines and red dashed lines represent the sub-bands for spin-up and spin-down electrons, respectively. Similar to the ZGNRs, the energy bands are spin-degenerate. In addition, four nearly flat bands e1-e4 exist in the vicinity of the Fermi energy, which are according to the four peaks p1-p4 in the DOS. Figure 3(c) shows the partial DOS (PDOS) of the zigzag chain 1/3, zigzag chain 2/4 and armchair chain 1/2, respectively. It can be seen that, due to the spatial symmetry, the PDOS of the zigzag chain 1 (2) is the same with the zigzag chain 3 (4), and the PDOS of the armchair chain 1 is the same with the armchair chain 2. Figure 3(d) shows the PDOS of the edge atoms and all atoms. The results of figures 3(c) and (d) indicate that the p orbitals of the zigzag chains contribute the most to the peaks p1-p4 in the DOS, thus the four nearly flat bands e1-e4 accommodate the localized edge states.

### 3.3. Magnetic properties of GCGNRs with single vacancy

Vacancy defects in ZGNRs and AGNRs have been studied for a long time \[16, 49\], however, there is a lack of research on the vacancy effects in CGNRs \[28\]. Without loss of generality, we choose one type of GCGNRs in figure 4. As a comparison, figure 4(a) illustrates the magnetic structures of an ideal GCGNR with the antiferromagnetic ground state. In addition, the spin population has a damped oscillation as a function of the...
nearest-neighbor index. Figure 4(c) shows one vacancy near the center of the nanoribbon. For simplicity, we call the three carbon atoms next to the missing one (denoted by A, B, C) the vacancy-nearest (V-N) carbon atoms. Different from the case in [28] where a new bond forms between two V-N carbon atoms and the third V-N carbon atom is passivated by one hydrogen atom, we passivate all three dangling bonds by hydrogen atoms near the vacancy. The spin-polarized calculations show that the spin-down electrons are localized around the V-N carbon atoms. Furthermore, we can see that the V-N carbon atoms and the edge atoms D, E, F, G belong to the same sublattice, so they host the localized states of the same spin species. So the natural question is: if the V-N carbon atoms and the ones at the zigzag chains belong to different sublattices, what will happen? To figure out the answer, we consider the case where the vacancy is near the edge of the nanoribbon in figure 4(e). It is clearly that, since the V-N carbon atoms A, B, C and the edge atoms D, E, F, G belong to the different sublattices, the localizations are both weaken around them. Meanwhile, because the atoms H, I, J, K in the lower edge share the same sublattice with the V-N carbon atoms, the localized states of the lower edge are enhanced. This suggests that there is close correlation of the spin localized states between the V-N carbon atoms and the edge atoms. Figure 4(b), (d) and (f) illustrate the energy bands of the GCGNRs in (a), (c) and (e), respectively. Figure 4(b)

Figure 3. (a) Spin-up (red) and spin-down electrons (blue) localize in the GCGNR (b) The energy bands and DOS of the GCGNR in (a). The four peaks p1-p4 in DOS are according to the four nearly flat bands e1-e4. (c) PDOS of the zigzag chain 1/3, zigzag chain 2/4 and armchair chain 1/2, respectively. (d) Comparison of PDOS between the edge atoms and all atoms.
means that the ideal GCGNR in (a) is spin-degenerate. Figure 4(d) and (f) show an occupied sub-band for spin-down electrons at the Fermi energy and an unoccupied sub-band for spin-up electron above the Fermi energy, which indicates that the total spin of the system is 1/2.

It should be pointed out that the famous Lieb’s theorem [44] can be used to analyse the magnetic ground states of the bipartite system. The Lieb’s theorem claims that, considering the electron-electron repulsive interaction, the ground state of a bipartite system at half-filling has the total spin:

$$ S = \frac{1}{2} |N_A - N_B| = \frac{1}{2} |N|, $$

where $N_A$ and $N_B$ denote the occupation number of the sublattices A and B, respectively. It should be pointed out that the result of equation (1) is also obtained from the ground state of large alternant organic molecules with conjugated bonds historically [30]. As a result, the imbalance between the two sublattices is the origin of magnetism in graphene. It should also be noted that antiferromagnetic ground state can exist when $S = 0$ in equation (1).

As shown in figures 4(c) and (e), the lattice distortion caused by the vacancy defect is not significant and the effect of the dangling bonds has been removed by the hydrogen atoms, so the Lieb’s theorem can be used to

Figure 4. (a), (c), (e) show the vacancy defect can enhance or weaken the localized edge states, depending on the V-N carbon atoms and the edge atoms occupy the same sublattice or not. (b), (d) and (f) illustrate the energy bands of the GCGNRs in (a), (c) and (e), respectively.
explain the magnetic ground states of our system. Because we only remove one carbon atom, the sublattice imbalance $|N_l| = 1$. According to equation (1), the total spin of the system is $1/2$, which directly leads to a local magnetic moment of $1\mu B$. This result is in agreement with our calculations.

As we know, zigzag edge states only survive when the length of the zigzag chains is long enough [11]. In GCGNRs, there may be many short zigzag and armchair chains in the edges. If one zigzag chain is very short, there will be no edge states. However, according to the above analysis, we know that there is a close correlation of the localized states between the V-N carbon atoms and the edge atoms. So one interesting question arise: can we use the vacancy in the bulk to induce or enhance the magnetic states in the edge?

To figure out the answer, we build GCGNRs in figures 5(a), (c), (e). For the ideal GCGNR without vacancy defects shown in figure 5(a), there is no edge state due to the short lengths of the zigzag chains, and the system is spin-degenerate which can be seen from the corresponding band structure in figure 5(b). In figure 5(c), a vacancy defect is introduced near the upper edge, thus localized spin states emerge around the V-N carbon atoms. Because the edge atoms D, E, F, G occupy the same sublattice with the V-N carbon atoms, localized edge states miraculously appeared due to the close correlation of the vacancy and edge. Furthermore, since the atoms H, I, J, K in the lower edges occupy the different sublattices with the V-N carbon atoms, there is still no edge states around them. Then we change the location of vacancy in figure 5(e), and in this case, the atoms H, I, J, K and the V-N carbon atoms share the same sublattice. As a result, except for the weak localization nearby in the upper edge, the magnetic states also survive in the lower edge and spread over the nanoribbon. Consequently, it reveals that the magnetic states in the edge can be induced or enhanced by the bulk vacancies. In other words, we may

![Figure 5](image-url)
obtain the desired magnetic edge states by tuning the vacancies or adatoms in the bulk. Furthermore, similar to the band structures in figure 4(d) and (f), figures 5(d) and (f) also illustrate an occupied spin-down sub-band and an unoccupied spin-up sub-band near the Fermi energy. This suggests that the total spin is 1/2, which is in agreement with our spin-polarized calculations in which we get a local magnetic moment of 1 μB.

4. Conclusion

In this work, we study the unzipping rules of SWCNTs, and obtain the GCGNRs which possess several zigzag and armchair chains. By using the first-principles calculations, the magnetic states of GCGNRs have been investigated. We find three major properties of the GCGNRs: (i), several zigzag and armchair chains with different orientations coexist in one edge of the GCGNRs. The flat edges are energetic favorable in ideal unzipping processes. However, due to the effects of adatoms, inevitable impurities, etc., the unzipping paths may be complex, leading to the generalized chiral edges. (ii), near the junctions of neighbouring zigzag chains with different orientations, the antiferromagnetic order in one edge and the ferromagnetic order between different edges can emerge. This is totally different from the well-known edge states of ZGNRs in which the ferromagnetic order exits in one edge and the antiferromagnetic order exits between different edges. (iii), the magnetism of GCGNRs can be explained by the Lieb’s theorem, and the single vacancy defects may induce or enhance the edge states of the GCGNRs. (iv), the single vacancy defects could produce localized states near them and provide local magnetic moments due to the Lieb’s theorem. More importantly, the localized states belong to the vacancies and edges can interplay with each other. Depending on the sublattice type of the vacancy and edge atoms, the localized states near the vacancy may induce or enhance the edge states. As a result, the desired magnetic edge states may be obtained by tuning the vacancies in the bulk.

GNRs, which possess excellent electronic properties, are potential for both transistors and interconnects. We have mentioned that GCGNRs could appear in the synthesis processes of nanoribbons, thus effects caused by generalized chiral edges are significant in GNR circuits. Especially, whether the GCGNRs are metals or semiconductors is crucial. Moreover, GNRs have been studied for building blocks in the circuit level, and generalized chiral edges may be common in such block structures. Furthermore, the novel magnetic properties of edge states mentioned in the paper could also influence the performances of the circuits. On the whole, this is an interesting topic, and more details can be studied in the future.

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References

[1] Novoselov K S, Geim A K, Morozov S V, Jiang D, Katsnelson M I, Grigorieva I V, Dubonos S V and Firsov A A 2005 Two-dimensional gas of massless Dirac fermions in graphene Nature 438 197–200
[2] Geim A K and Novoselov K S 2007 The rise of graphene Nature Mater. 6 183–91
[3] Neto A H C, Guinea F, Peres N M R, Novoselov K S and Geim A K 2009 The electronic properties of graphene Rev. Mod. Phys. 81 109–62
[4] Wu X J and Zeng X C 2008 Sawtooth-like graphene nanoribbon Nano Res. 1 40–5
[5] Yeo P S E, Loh K P and Gan C K 2012 Strain dependence of the heat transport properties of graphene nanoribbons Nanotechnology 23 495702
[6] Nakada K, Fujita M, Dresselhaus G and Dresselhaus M S 1996 Edge state in graphene ribbons: Nanometer size effect and edge shape dependence Phys. Rev. B 54 17954–61
[7] Wu M H, Wu X J, Pei Y and Zeng X C 2011 Inorganic nanoribbons with unpassivated zigzag edges: half metallicity and edge reconstruction Nano Res. 4 233–9
[8] Klein DJ 1994 Graphitic polymer strips with edge states Chem. Phys. Lett. 217 261–5
[9] Kusakabe K and Maruyama M 2003 Magnetic nanographite Phys. Rev. B 67 092406
[10] Suenaga K and Kohsaka M 2010 Atom-by-atom spectroscopy at graphene edge Nature 468 1088–90
[11] Bao Z Q, Shi J J and Zhang M 2013 Ferromagnetism, adatom effect, and edge reconstruction induced by klein boundary in graphene nanoribbons J. Appl. Phys. 113 194302
[12] Wagner P, Ivanovskaya V V, Melle-Franco M, Humbert B, Adjizian J J, Briddon P R and Ewels C P 2013 Stable hydrogenated graphene edge types: normal and reconstructed klein edges Phys. Rev. B 88 094106
[13] Tabarreaa A, Shadaloua S and Song J-H 2015 Mechanical properties of graphene nanoribbons with disordered edges Comp. Mater. Sci. 96 10–6

[14] Koskineena P, Malola S and Häkkinnen H 2008 Self-passivating edge reconstructions of graphene Phys. Rev. Lett. 101 115502

[15] Huang B, Liu M, Su N, Wu J, Duan W, Gu B-L and Liu F 2009 Quantum manifestations of graphene edge stress and edge instability: a first-principles study Phys. Rev. Lett. 102 166404

[16] Dutta S and Pati S K 2010 Edge reconstructions induce magnetic and metallic behavior in zigzag graphene nanoribbons Carbon 48 4409–15

[17] Pincak R, Smoluchawa J and Osipov V A 2015 Electronic states of zigzag graphene nanoribbons with edges reconstructed with topological defects Physica B 475 61–5

[18] Park S-Y, Moon K and Rhim J-W 2015 Intrinsic half-metallicity of the Stone-Wales edge reconstructed graphene nanoribbons Phys Status Solidi B 252 339–45

[19] Yazeyev O V, Capaz R B and Louie S G 2011 Theory of magnetic edge states in chiral graphene nanoribbons Phys. Rev. B 84 115406

[20] Li X-F, Wang L-L, Chen K-Q and Luo Y 2012 Strong current polarization and negative differential resistance in chiral graphene nanoribbons with reconstructed (2,1)-edges Appl. Phys. Lett. 101 073101

[21] Tao C et al 2011 Spatially resolving edge states of chiral graphene nanoribbons Nature Phys. 7 616–20

[22] Goler M, Lang Y C and Wessel S 2013 Quantum Monte Carlo studies of edge magnetism in chiral graphene nanoribbons Phys. Rev. B 87 155441

[23] Jiang Z and Song Y 2015 Band gap oscillation and novel transport property in ultrathin chiral graphene nanoribbons Physica B 464 61–7

[24] Lebedeva I V, Popov A M, Knizhnik A A, Khlobystov A N and Potapkin B V 2012 Chiral graphene nanoribbon inside a carbon nanotube: ab initio study Nanoscale 4 4522–9

[25] Chang P-H and Nikolic B K 2012 Edge currents and nanopore arrays in zigzag and chiral graphene nanoribbons as a route toward high-ZT thermoelectrics Phys. Rev. B 86 041406

[26] Carvalho R R, Warnes J H and Lewenkopf C H 2014 Edge magnetization and local density of states in chiral graphene nanoribbons Phys. Rev. B 89 245444

[27] Tan X-D, Kang X-B, Zhao L-M, Zhang J-J and Hao H-S 2019 Quantum dissonance in chiral graphene nanoribbons J. Phys.: Condens. Matter 31 205602

[28] Sahni Z and Berber S 2020 Monovacancy in achiral and chiral graphene nanoribbons Computational Condensed Matter 23 e00471

[29] Bai J, Duan X and Huang Y 2009 Rational fabrication of graphene nanoribbons using a nanowire etch mask Nano Lett. 9 2083–7

[30] Wang X and Dai H 2010 Etching and narrowing of graphene from the edges Nat. Chem. 2 661–5

[31] Abramova V, Slesarev A S and Tour J M 2015 Meniscus-mask lithography for fabrication of narrow nanowires Nano Lett. 15 2933–7

[32] Cai J et al 2010 Atomically precise bottom-up fabrication of graphene nanoribbons Nature 466 470–3

[33] Vo T H, Shkhiriev M, Kunkel D A, Morton M D, Berglund E, Kong L, Wilson P M, Dowben P A, Enders A and Sinitskii A 2014 Large-scale solution synthesis of narrow graphene nanoribbons Nat. Commun. 5 3189

[34] Warner J H, Rümmeli M H, Ge L, Gemming T, Montanari B, Harrison N M, Büchner B and Briggs G A D 2009 Structural transformations in graphene studied with high spatial and temporal resolution Nat. Nanotechnol. 4 500–4

[35] Börnert F, Fu L, Gorantla S, Knupfer M, Büchner B and Rümmeli M H 2012 Programmable sub-nanometer sculpting of graphene with electron beams ACS Nano 6 10327–34

[36] Kosynkin DV, Higginbotham AL, Sinitskii A, Lomeda JR, Dimiev A, Price BK and Tour JM 2009 Longitudinal unzipping of carbon nanotubes to form graphene nanoribbons Nature 458 872–7

[37] Jiao L, Zhang L, Wang X, Diankov G and Dai H 2009 Narrow graphene nanoribbons from carbon nanotubes Nature 458 877–80

[38] Kosynkin DV, Lu W, Sinitskii A, Perea G, Sun Z and Tour JM 2011 Highly conductive graphene nanoribbons by longitudinal splitting of carbon nanotubes using potassium vapor ACS Nano 5 968–74

[39] Kumar P, Panchakarla I S and Rao C N R 2011 Laser-induced unzipping of carbon nanotubes to yield graphene nanoribbons Nanoscale 3 2127–9

[40] Genorio B, Lu W, Dimiev A M, Zhu Y, Raji A R O, Novosel B, Alemany I B and Tour J M 2012 In situ intercalation replacement and selective functionalization of graphene nanoribbon stacks ACS Nano 6 4231–40

[41] Song Y, Long M, Liu G, Gao S, Zhu C, Wei X, Geng X, Sun M, Yang C and Lu L 2013 Electronic transport properties of graphene nanoribbon arrays fabricated by unzipping aligned nanotubes Phys. Rev. B 87 165404

[42] Xiao B, Li X, Li X, Wang B, Langford C, Li R and Sun X 2014 Graphene nanoribbons derived from the unzipping of carbon nanotubes: Controlled synthesis and superior lithium storage performance J. Phys. Chem. C 118 881–90

[43] Dimiev A M, Khananov A, Kiamov I V A, Shukhina K and Tour J M 2018 Revisiting the mechanism of oxidative unzipping of multilayer carbon nanotubes to graphene nanoribbons ACS Nano 12 3985–93

[44] Lieb E H 1989 Two theorems on the Hubbard model Phys. Rev. Lett. 62 1201–4

[45] Segall M D, Lindan P J D, Probert M J, Hasnip P J, Clark S J and Payne M C 2002 First-principles simulation: ideas, illustrations and the CASTEP code J. Phys. Condens. Matter 14 2717–44

[46] Vanderbilt D 1990 Soft self-consistent pseudopotentials in a generalized eigenvalue formalism Phys. Rev. B 41 7892–5

[47] Perdew J P, Burke K and Ernzerhof M 1996 Generalized gradient approximation made simple Phys. Rev. Lett. 77 3865–8

[48] Monkhorst H J and Pack J D 1976 Special points for Brillouin-zone integrations Phys. Rev. B 13 5188–92

[49] Akhmerov A R and Beenaker C W J 2008 Boundary conditions for Dirac fermions on a terminated honeycomb lattice Physical Review B 77 085423

[50] Ovchinnikov A A 1978 Multiplicity of the ground state of large alternant organic molecules with conjugated bonds Theoret. Claim. Acta (Berl.) 47 297–304