

Corollaries of Deforming Graphene Nanosheets Using Isopropyl Alcohol

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Abstract

The electrons in graphene can be manipulated by fabricating suitable deformation of its crystal lattice such that it imitates electric and magnetic fields. However, its Young's modulus of 1TPa makes it difficult to deform graphene permanently. The energy, time and sophistication involved in deforming and machining graphene to required specification is high. This research focuses on a technique that allows graphene to be deformed successfully with minimal effort. In this novel method, the advantages pertinent to other deformed graphene like quantum dots (GQD) and nanoribbons (GNR) are adapted and accomplished by deforming planar graphene sheets without complexity. Graphene's hydrosensitivity and allergy to temperature depression were recruited to execute this task alongside a simple working tool.

Keyword: deformation, graphene, freezing point depressants, hydrosensitivity

1. Introduction

What plastic was in the 20th century is what graphene will be in the 21st century [1]. This material has more than ten times better electronic conduction properties than silicon such that it is considered as an ideal replacement for silicon [2]. Graphene can deliver electronic devices much smaller as well as sustain an electron transport speed that can reach 1/300 of the speed of light [3]. Graphene is able to absorb a wide range of photo frequencies as against other semiconductors [4, 5], and it is more efficient than their silicon analogs². The proficiencies of graphene are too numerous to mention.

However, most of these capabilities are better harnessed when graphene is machined. Theoretical works advocate

that strain engineering of graphene can facilitate dirac cone merging, band gap opening and pseudo magnetic field creation [6]. Graphene is made luminescent by inducing a band gap using two major paths. One is by cutting it into ribbons and quantum dots and the other is by chemical or physical treatments, to reduce the connectivity of the π electrons network [7]. Carbon nanostructures like nanotubes, fullerenes and graphenes are given high consideration when properties such as such as good thermal conductivity [8], corrosion resistance [9], good electronic property [10], magnetism¹, etc. are of importance. Other useful applications of deformed graphene include their resourcefulness in optical devices [11], solar cell technologies [12, 13], water

filtration [14], and sensors [15] and so on.

It has been discovered that superconductivity was imminent when twisting bilayer graphene at a certain angle. To achieve this, the two hexagonal nets were twisted against each other at a 1.1 degree angle and a critical temperature T_C of 1.7 K¹⁶.

Pristine carbon nanostructures are difficult to fabricate through conventional synthesis methods due to very high crystalline temperature which are usually above 1500°C¹⁷. Furthermore, their high modulus of elasticity of 1TPa compounds the problem. The electronic properties of graphene are vast and these properties are influenced by the shape, positioning and constraints of graphene sheets [18]. A graphene sheet is soft and flexible but deforming graphene into various shapes is difficult [19]. Obtaining a scroll, for instance, involves a large energy barrier due to the bending rigidity and this has to be subdued [20].

Folded graphene has displayed novel electrical and mechanical properties unequalled by pristine graphene [21], which implies that morphology of graphene adds the dimensionality of design space to modify its properties [22].

Graphene can be deformed, especially, in the direction that is out of the plane which is where the restitution forces vanish. Such deformations join directly

to the electrons, making graphene a remarkable case of a metallic membrane [18].

In the formation of graphene nanoscrolls, the DFT of rolling energy against rolling steps presents a non-trivial deformation of the scroll. By proper doping, the scrolling energy can also be reduced. Sharifi et al [23], alleges that, with doping, the rolling energy of a GNS can be reduced to less than its value of 5.5 eV to as low as 2eV (2-M-N-rGOx) or less. In contrast, the resultant sheet will no longer be pristine graphene and could lack some basic properties of original graphene [24].

Graphene has become a promising material for future electronics but one problem is the lack of a gap in its energy spectrum [25]. This gap is characteristic of silicon and other materials currently used by the semiconductor industry [26]. It is a major component of solar cell technology. Guinea et al [27], report that the graphene material is known to be flexible and can be stretched by as much as 20% without prompting defects. If forces are applied along three main directions in graphene's honeycomb crystal lattice, a semiconducting gap opens up which is large enough for electronic applications. Also, one way to make a uniform pseudo (virtual) magnetic field is to stretch a flat graphene sheet in three different directions, but this is technically challenging. However, Zhu et al [28] conceived that pulling along just one axis is enough. Strained

graphene allow electrons move as if in a strong magnetic field. Pseudomagnetic effect opens up new possibilities in graphene electronics. A new theoretical study describes how to shape a flat graphene ribbon so that pulling on its ends produces a uniform pseudomagnetic field. In 2009, it was discovered that stretching the 2D lattice of graphene into a triangular bubble shape initiated the conduction electrons to turn in a circular array just like electrons in a magnetic field. These pseudo magnetic fields are intensely peaked having magnitudes as high as 300 tesla. If the induced fields are made to be more spatially uniform; valleytronics [27] is possible.

Patra et al, demonstrated by molecular dynamics (MD) simulations that water nanodroplets can trigger and guide the folding of planar graphene nanostructures. They forecasted that nanodroplets can encourage rapid bending, folding, sliding, rolling, and zipping of the planar nanostructures, which result in the assembly of nano scale sandwiches, capsules, knots, and rings [29]. Xie et al [30], reported that by immersing mechanically exfoliated graphene in isopropanol/ water solution (IPA/ H₂O), graphene nanoscrolls (GNS) was successfully synthesized by a self-scrolling process after 5 minutes of immersion. They also reported that by adjusting the molality of the solution, structures such as folds and bends were obtainable.

Li et al [31] explained that the production of large quantities of GNS was accomplished by applying mild synthesis conditions to chemical vapour deposited (CVD) graphene. A CVD grown graphene sample on SiO₂ (0.5 cm × 0.5 cm) was placed in a glass vial containing 1/1 (v/v) methanoic acid/water (HCOOH/H₂O) mixture, such that the graphene sample was completely immersed in the aqueous HCOOH mixture at 45°C. After 24 hours GNS was self-fabricated from this treatment.

Mirsaidov et al [32] explained that scroll-shaped Nano channels are formed in the presence of the water layer between two graphene films. He explains further that water initiates scrolling of detached fragments of graphene from an etched copper substrate. GNS was formed after this hydrophobic process.

The four works, mentioned in the last two paragraphs above, are similar in that all the working fluid used were freezing point depressants. The difference is in the type of working fluid used (isopropyl alcohol, methanoic acid and water). While Xie et al [30] believe that the IPA was responsible for the scrolling, Mirsaidov et al [32] believe it was water. He explains that in the absence of water between two graphene layers there were no nanochannels, which indicates that the presence of the water between two graphene layers was needed for scrolling the graphene. Li et al [31] said that the aqueous HCOOH

mixture was responsible for scrolling. However, all these works agreed that adjusting the molality of the mixtures with water determined the outcome of the scroll formed.

Water has a remarkable effect on graphene. Graphene's hydrophobicity largely depends on the thickness; single layer graphene being considerably more hydrophilic than multiple layers of graphene³³. Algara-Siller et al [34] reported that when water is sandwiched between two sheets of graphene and pressure is applied; the part of the water molecule not lost forms a rare square ice lattice crystalline phase. A strong adhesion between the graphene sheets by way of Van Der waal forces that form the nanocapillary are responsible for some of the pressure which could be as high as 1 GPa. This phenomena appears to be an important factor in making water crystallize into ice at room temperature; an example of hydrophobic restriction [32].

This feat was confirmed by Bampoulis et al [35] when water was dropped between a sheet of molybdenum disulphide (MoS_2) and graphene.

This present research proposes a method which allows graphene sheets to be deformed into other shapes while in isopropyl alcohol (IPA) /water ($\text{CH}_3\text{CHOHCH}_3/\text{H}_2\text{O}$) solution easily. This is a case where resistance to mechanical flow is minimized because of graphene's hydrosensitivity. As such, stretching it or performing any other

deformation operation will be easier and cheaper.

2. Experiment

Firstly, graphite rods from 8B marker were crushed and graphene was synthesised by mechanical exfoliation³⁰ out of the crushed graphite rods.

Preliminary investigations were carried out using SEM and EDS to ascertain the elemental state of the graphene sample before the deformation process began. When this preliminary result was compared with the final result obtained after deformation; appropriate judgements were concluded for graphene's performance.

For this purpose, the raw exfoliated samples were loaded unto the Zeiss EVO LS 10 SEM/EDS and preliminary analysis of graphene structures began. Scanning electron microscopy (SEM) provided a picture of the morphology and structure of grapheme [36] while EDS provided the elemental composition survey, respectively. The results obtained from this preliminary investigation are displayed in table i and figure 1 respectively.

The sample, placed on SiO_2 substrate, was thereafter removed from the SEM compartment and immersed in IPA/ H_2O solution in the ratio of 1:3 for 5 minutes. This solution encompassing the

graphene was stirred in a zigzag manner, thoroughly, with a pin. This was done to observe whether the outcome will show some modifications in the shape of the GNS and other graphene structures that will emerge. The sample was removed and left to dry for about two hours at 32°C. The choice of sample participants was not limited to single layer graphene but a few multiple layered graphene sheets (which is typical of mechanically exfoliated graphene samples) were present in the sample.

The final investigations followed this manual deformation process. Again, the sample was loaded unto the SEM for surface morphology and X-ray analysis. The SEM/EDS results obtained from this final experiment are displayed in table ii and figures 2-5 respectively.

3. RESULTS

3.1 Preliminary investigation

Table i: EDS composition of area (1404)

eZAF Smart Quant Results										
Creation D #####										
Project	New Sample Area 1404 Selected Area 1									
Specimen										
Area										
Sampling Region										
kV	15									
Live Time	30									
Amp Time	3.84									
Takeoff Ar	35.18									
Resolution	125.625									
Element	Weight %	Atomic %	Error %	Net Int.	K Ratio	Z	R	A	F	
C K	72.14	87.19	34.76	1415.03	0.248	1.0481	0.9746	0.328		1
O K	1.09	0.99	31.16	25.6	0.0018	0.9974	0.9943	0.1693		1
Al K	4.72	2.54	3.97	894.69	0.0361	0.8793	1.0301	0.8589		1.0119
Si K	12.82	6.63	2.89	2404.33	0.1032	0.8974	1.0357	0.892		1.0052
Ca K	2.49	0.9	7.58	222.74	0.0218	0.8409	1.0611	0.9995		1.0406
Fe K	6.73	1.75	6.91	218.24	0.0541	0.7409	1.067	1.0098		1.0742

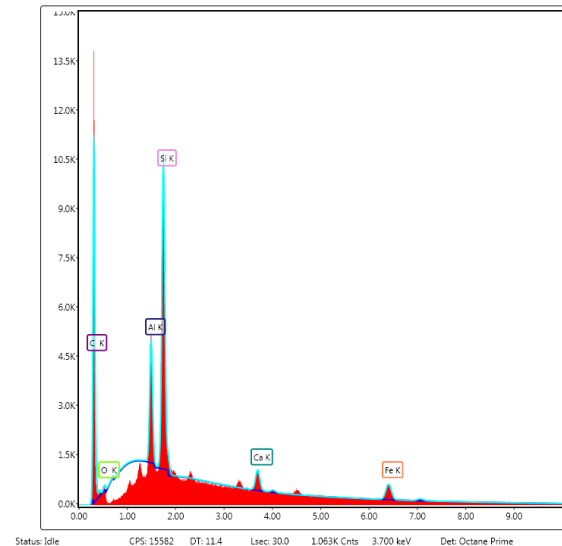


Figure 1: Spectrum of Area (1404)

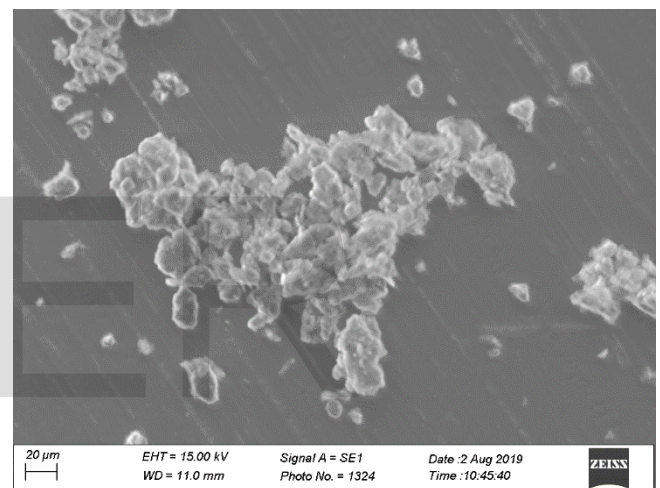


Figure 2: SEM Imaging of 1407

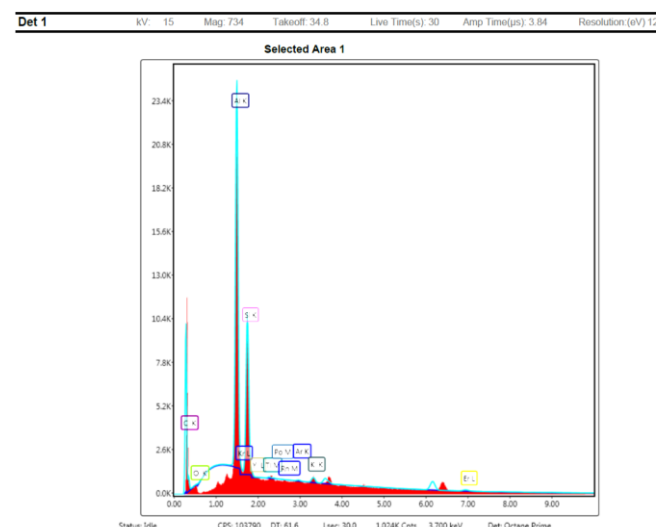


Figure 3: EDS Spectrum of 1407

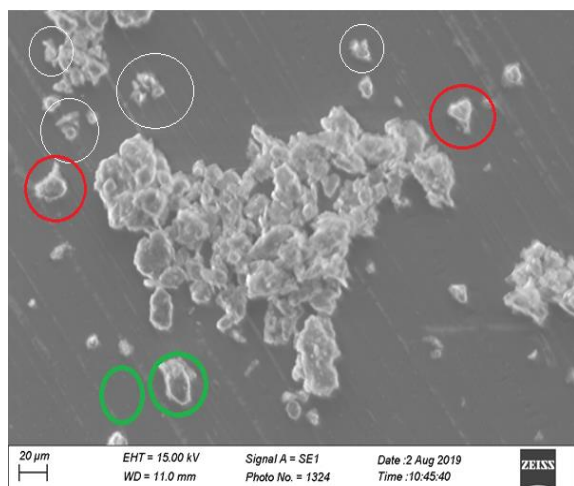


Figure 4: SEM Imaging of 1407 (edited)

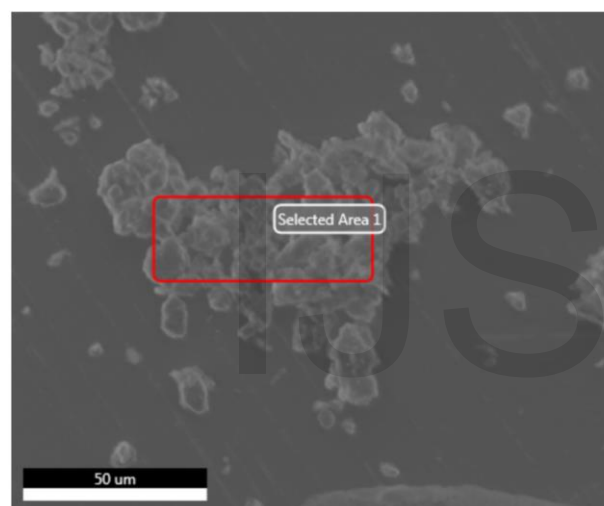


Figure 5: Area selected for EDS

Table ii: EDS Composition of GNS 1407

eZAF Smart Quant Results												
Creation D #####												
Project New Sample Area 1407 Selected Area 1												
Specimen												
Area												
Sampling Region												
kV	15											
Live Time	30											
Amp Time	3.84											
Takeoff Ar	34.83											
Resolution	125.625											
Element	Weight %	Atomic %	Error %	Net Int.	K Ratio	Z	R	A	F			
C K	67.03	83.4	9.3	1424.49	0.1745	1.0587	0.972	0.246	1			
O K	0	0	99.99	0	0	1.0077	0.9919	0.1745	1			
Al K	19.39	10.74	2.68	5474.55	0.155	0.8886	1.0282	0.8941	1.0063			
Kr L	0.38	0.07	20.25	44.98	0.0025	0.6806	1.1999	0.9478	1.0108			
Si K	9.86	5.25	3.83	2386.98	0.0719	0.9069	1.0339	0.8006	1.0035			
Y L	0.04	0.01	67.88	4.2	0.0003	0.6865	1.2121	0.9309	1.011			
Ti M	0.23	0.02	58.41	22.37	0.0019	0.5612	1.4104	1.1884	1.2522			
Po M	0	0	99.99	0	0	0.5658	1.4169	1.1739	1.3209			
Rn M	0	0	31.29	0	0	0.5428	1.4189	1.1776	1.3911			
Ar K	0.13	0.05	56.49	20.53	0.001	0.7827	1.0527	0.9492	1.0157			
K K	0.7	0.27	8.42	104.45	0.0058	0.8358	1.0565	0.9709	1.0184			
Er L	2.24	0.2	11.61	39.06	0.0145	0.5288	1.1566	1.0469	1.1668			

4. DISCUSSION

4.1 Preliminary Investigation

From the analysis of preliminary investigations on the graphene sample (area 1404) using SEM and EDS (see table i), it is very obvious that the graphene sheets were bordered with hetero-nano particles of iron (Fe), calcium (Ca), aluminium (Al) and silicon (Si). This outcome was not surprising because all these elements make up clay. The average clay sample contain these elements in the form of FeO, CaO, Al₂O₃ and SiO₂ respectively³⁷. Clay is a major component of the graphite marker. It is used to bind graphite together. Testing conducted on various locations in the sample confirmed the same result for graphene and graphene oxide with slight distortions in the percentages.

4.2 Final Investigation

Graphene Sample Area 1407 (see figure 2). Sharp indentations, incomplete scrolls, closed ended graphene, super structures and folds marred the sample; meaning that while in IPA/water solution, the rigorous stir helped to deform, shape, bend and strain graphene. This process did not require much sophistication to activate other than the manual zigzag stirring using the pin. Figure 4 shows the SEM image of folded graphene structures; the white circles present graphene structures with sharp depressions and bends while the red circle present a closed ended scroll. Several uncompleted scrolls were seen

in Figure 4. A number of graphene structures synthesized by this rigorous stir presented some bending tendencies. Despite the disparity in size and shape, these graphene structures showed several similarities. The graphene sample was examined as a whole and their collective property was analysed. However, individual sheets and how they were deformed were analysed in some cases.

Hooke's law holds that for elastic materials like graphene, stress is proportional to strain with the Young's modulus being constant [38].

Guinea et al [39] expounds that it is possible to create strain in graphene by bending or deforming it. They further explain that in-plane bending of graphene ribbons should lead to strong pseudo magnetic effects. Lu et al [40] explains that bending and in-plane strain occurs together and even proposed an equation to link them up in graphene. In fact, self-deformation such as scrolling in graphene is the result of surface strain of its sheets [30]. Pristine graphene has zero band gap [41] but according to Guinea et al²⁷, an energy band gap for graphene is realized when the graphene is stretched or strained. This band gap is the major phenomena that justify the use of semiconductors like silicon in solar cells²⁶. Deformation is a means by which this band gap is realised by strain or stretch. The odd shapes like bends, scrolls and deformations produced in this present research imply that some in-plane strain had been inputted onto these

sheets as a result of bending and other stresses formed; inferring that the insertion of a band gap is likely. Nevius et al [2] explains that one way of introducing band gap is by deforming graphene into odd shapes, such as ribbons from which band gaps have obtained. This present study has achieved this by deforming graphene in IPA/H₂O solution.

One exceptional feature of this graphene sample was its glowing appearance. According to Bonaccorso et al [7], graphene is also made luminescent by inducing a band gap. The sheets emitted a high luminosity. This illumination helped in the easy identification of some bent and folded graphene structures identified in white circles on figure 4. The red circle in figure 4 shows scroll structure that is tapered into a cone (i.e. closed ended). This shape is made visible because of the luminosity of the deformed graphene structures (which are generally referred to as graphene material in this work) on a dark background. Luminescence is the emission of radiation due to an electron in an excited energy state falling to a lower energy state^{42;43}. This is the event of the rigorous stir in IPA/water solution. The stir permits electrons in the sheets to experience maximum excitation. This luminosity confirms that the action carried out in this work actually succeeded in performing band gap engineering on graphene sheets.

From figure 4, it is visible that the graphene materials are transparent.

Using optical contrast in comparing the background of the area behind the sheets and that on the empty space (see the green circles), it can easily be concluded that the graphene material is transparent; especially with the illuminated sample. Jiang et al [41] maintains that graphene is transparent. As such, this is a further justification that the sample is graphene.

Upon sighting the EDS results for sample 1407 and comparing the results before and after the experiments (see table i and ii), it was obvious that table ii was laden with several elements stuck onto the graphene material. This is a special case of super adhesion facilitated by charged graphene structures. Polonium (Po) and Radon (Rn) were among the nine elements picked up by this graphene material. Deformed graphene like GNS are known to be adhesive up to 1.72Jm^{-2} of adhesion energies [31]. Some earth samples were tested in the SEM prior to this testing and sample 1407 picked all these chemical elements from the remnant of these earth materials. Rao et al¹ suggest that zigzag edges may responsible for the *magnetic properties of graphene*. Li et al³¹ clarifies that the superior adhesion mechanism could be related to the strain in the graphene layer. Guinea et al³⁹ further explain that in-plane bending of graphene ribbons should lead to strong pseudo magnetic effects. These conditions were satisfied in this work and they are the likely reason for the kind of adhesion observed in this present study.

A major corollary of deforming graphene with IPA/ water is the apparent excitation of electrons of graphene sheets. This is an indication that this working fluid has a remarkable effect on graphite and its substructures on the whole since graphite is the product of multiple graphene sheets stacked up together⁴⁴.

The ease of formation of the scroll and other super structures in this work imply that the cost of initiating deformation to graphene in IPA/water is cheaper than most methods mentioned in literature²⁸. Further stretching with the use of sophisticated tools will also be supported when graphene is immersed in IPA/water solution.

Resistance to deformation or Young's modulus levels up to 1TPa in graphene and this is highly suppressed in the presence of IPA/water which take advantage of graphene's hydrosensitivity (phobic and philic) [33] to influence the shape and flexural swings of graphene sheets. Thus, making graphene behave either like a non-metal or like a semi metal as it is called⁴⁵. Solids maintain their shape in the presence of stress up to an extent³⁸ but graphene flows quite easily in the presence of IPA/water, mimicking a non-metal. Hence, the usual alias of graphene as semi metal.

5. COROLLARY AND CONCLUSION

IPA/water is a great machining tool for graphene. A lot of machining processes involving graphene are cheaper when the graphene sample is immersed in IPA/water. It does more than just scrolling operation on graphene; it is a valuable nano-working fluid. Adjusting the molality of the solution determines the outcome of the shape formed. For all self-scrolling theories mentioned; water plays the role of adjusting the molality of solutes [30'32].

The properties of the deformed graphene material described in this work will provide solutions in the decontamination of a polluted site as pollutant population will adhere to the graphene material thereby, ridding the site of such contaminants when the procedure is performed repeatedly [51].

This graphene material will also serve as a major sensor for determining the constituents of elements in a selected site and it will show the degree of contamination if it is combined with a spectrometer; thereby, performing the role of a contamination sensor [51]. This revelation will be very useful in the production of bio-sensors and chemical sensors.

This study also reveals that graphene is allergic to some materials, especially freezing point depressant like HCOOH [31], IPA [30], H₂O or any process involving freezing point depression and as such graphene will physically react to

their stimuli in a way that would influence its orientation and shape thus affecting its electron dispersion and other properties. Alcohol [46'46], water [48'49] and methanoic acid [50] are freezing point depressants. Xie et al [30] reported that by immersing mechanically exfoliated graphene in isopropanol/ water solution (IPA/ H₂O), it was possible to self-scroll graphene. Patra et al²⁹ showed that H₂O alone could achieve the same feat using MD simulation. Similarly, Li et al [31] proved that methanoic acid/water (HCOOH/H₂O) solution could self-scroll graphene whereas this present research has shown that when graphene is immersed in IPA/ H₂O solution and the set-up is stirred rigorously with an external force; several potentials other than scrolling are unlocked.

An explanation to this entire phenomena lie in the fact that graphene is sensitive to temperature depression. Any material that is capable of depressing or interfering with its temperature can stimulate it into deformation. Graphene's line of defence to allergy is to coil itself in a bid to reduce its surface area interfacing with the source of allergy, thus, resulting in scrolling. Other shape permutations are possible when an external force is applied or the molality of the solution is adjusted; thereby initiating bandgap, pseudomagnetic effects and luminosity, etc.

Most of the corollaries reported above will play out with most 2d materials.

6. Acknowledgments

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