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Review Article

Methods of inducing magnetism into graphene monolayer

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ABSTRACT

Graphene has made a revolution in the material industry after the demonstration of the ability to successful isolation of graphene thin layers in 2004, due to its exclusive characteristics of electronic, optical, and mechanical properties. However, ideal graphene has no net magnetic moment due to a delocalized π-bonding in nature and has a zero bandgap which limits its practical applications in the world. Nevertheless, many studies have shown that by inducing magnetism into graphene, the mentioned limit can be overpowered which enables to apply especially in spintronics and memory storage devices. Therefore, this review paper discusses four possible techniques in improving magnetic properties on graphene monolayer, which are surface doping with adatoms, vacancy creation, substitutional doping, and edge modification, by providing the experimental, theoretical evidence and as well as their limitations, future research, and applications.

Keywords: Graphene, Magnetic properties

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1. INTRODUCTION

Graphene is a monolayer sheet consisting of a tightly bounded 2D array of hexagonal honeycomb lattice formed by carbon (C) atoms, that was isolated experimentally in 2004 [1]. Literature defines a high possibility of integrating graphene into novel applications in diverse fields of electronics, energy conversion, and flexible information storage systems [2, 3] due to its outstanding and unique mechanical, electronic, magnetic, and optical properties [2]. Out of these, the magnetic properties of graphene are one of the very important research areas to understand its behaviour and mechanism, in order to develop graphene-based electronic and optoelectronic devices for various applications. As graphene has a long spin-relaxation time and ballistic transport characteristics [4], it can be specially used in spintronic devices [3]. However, the unique properties of the zero-band gap at the Dirac point and the nonmagnetic environment of non-defective graphene monolayer [1] have restricted the wide range of potential applications of it in the practical world.

The magnetic properties in the materials mainly rely on the magnetic moment resulting from the motion of the electrons. As the electron performs two kinds of rotations, the orbital moment and the spin emerged [4]. According to their bulk magnetic susceptibility, the materials can be classified as diamagnetic, paramagnetic, or ferromagnetic [4]. The electron spin magnetic moment is classified as the fundamental quantity, Bohr magneton (μ_B) [4].

$$\mu_{spin} = \frac{eh}{4\pi cm} = 0.927 \times 10^{-20}\, emu = \mu_B \qquad (1)$$

Where, *h* - Plank constant, *m* - electron mass, c-speed of light and, *e*- electron charge. The magnetic moment of the atom can be obtained by vector addition of all the electron moments, and the total magnetization of the graphene monolayer can be calculated from [4]:

$$M_{total} = \int \left(n_{up} - n_{down}\right) d^3r \qquad (2)$$

Where n_{up} and n_{down} are the number of spins in the up and down direction. However, spin polarization and magnetism cannot occur in graphene monolayer due to the absence of unpaired electrons [5, 6].

Most of the common magnetic materials have 3d/4f transition metals integrated into them in order to enhance the magnetic properties by s/p electrons [7], which can be adapted to enhance the magnetic properties of graphene. Nevertheless, when developing carbon-based magnetic materials, it is important to develop and understand the techniques to control the spin of s/p electrons [8]. As a solution, many studies have been triggered in this field as they have shown that the magnetic properties of graphene can be improved by manipulating electron structure [9]. Considering the difficulty of experimentally manipulating the electronic structure at the atomic level with the existing instruments and techniques [3, 10], only fewer literature on experimental studies can be observed on the topic of altering the magnetic properties of graphene monolayer [3, 10] but considering the future potential of utilizing graphene on spintronics and many other novel electronics devices, it is worthwhile to get a depth in knowledge of techniques on improving the magnetic properties of graphene. Therefore, this review paper highlights four possible techniques which are surface doping with adatoms [3], vacancy creation [3, 11], substitutional doping [3, 11] and, edge modifications[3] that can be employed for the task of altering the magnetism of monolayer graphene. It is also to note that the results presented in this article are based on these methods, most of the evidence on magnetic graphene can only be observed at low temperature rather than at room temperature which implies a limitation in inducing magnetization into graphene [9].

2. MAGNETISM-INDUCINGING METHODS

2.1 Surface Doping with Adatoms

When doped on the graphene surface with a selected dopant, the adatom can obstruct and alter the natural behaviour of graphene. The Adatom adsorption on graphene monolayer surface stimulates lattice deformation which leads to the electric dipole moment and the spin-polarized magnetic moment [6]. Figure 1(a) shows the three high symmetry sites (H, B and T) which are suitable for the adsorption of adatoms [3] and Figure 1(b) illustrates a lattice with adatoms on the H sites on a graphene monolayer.

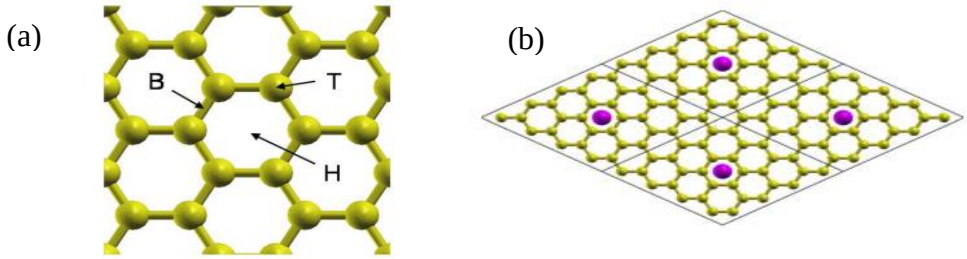


Figure 1: (a) The 3 different possible adsorption sites (H, B, and T) on graphene monolayer (b) Adatom on the H site on a graphene surface (yellow-carbon atom, pink-adatom) [12]

https://www.semanticscholar.org/paper/First-principles-study-of-metal-adatom-adsorption-Chan-Neaton/5f0c2fa22be5e7ee7614410abb22506273d6487a

Héctor González-Herrero *et al.*[5] have reported having observed a magnetic moment in graphene by doping the graphene surface (7×7 nm^2) with a single H atom. The Scanning Tunnelling Microscopy (STM) image of the observation is shown in Figure 2 [5] and they have suggested the reason behind this phenomenon as the changes in the hybridization of carbon from sp^2 to sp^3 and the removal of the p_z orbital. Furthermore, the induction of magnetic moment due to Co and Fe single adatoms adsorbed in graphene monolayer are been confirmed theoretically by the DFT calculation individually[12,13] which also has been supported by the experimental evidence[10, 14, 15]. After Fe is adsorbed on graphene, the predicted electronic configuration is $3d^74s^1$ [10] while for Co is $3d^84s^1$ [10]. Not only the monoatomic elements but diatomic compounds have also contributed to the magnetization of graphene. As for the Co-N (cobalt-nitrogen) adsorption, a saturation magnetization of 0.11 emug^{-1} at 300K and 0.73 emug^{-1} at 5 K have been reported experimentally [16]. According to the above literature, the reason behind the induction of magnetism in the above cases is the p_z delocalization of N and C atoms and the hybridization between the *d* electrons of the Co and Fe atom, in respective cases [16].

The asymmetrical band diagram and Density of States (DOS) indicate the induced magnetism in the graphene monolayer. Figure [3, 6] illustrates such an asymmetric condition caused by adsorbing Mn [6 ,17]. Moreover, the adsorption of Ca[6], Ba[6] and Sr[6] atoms individually also have shown non-zero magnetic moments theoretically according to the asymmetric conditions of the band diagram and DOS.

In contrast, not all the elements will trigger the induction of magnetic moment. The adsorption of Ni adatom on monolayer graphene has not shown any magnetic moment

neither theoretically [10, 13] nor experimentally [10, 14]. According to the X-ray Magnetic Circular Dichroism (XMCD) measurements and X-ray Absorption Spectra (XAS)[10] which had been carried out in experimental studies, adsorption of Ni has not displayed any magnetic moment as shown in Figure 4. According to the observations of Co and Fe adsorption [10,] the predicted electron configuration for Ni after the adsorption should be $3d^9 4s^1$ [10]. However, the Ni-graphene system has not shown any magnetic moment. Thomas Eelbo *et al.* [10] suggested that the reason for the non-magnetism of the Ni-Graphene system is due to the conversion of electron configuration in the Ni monomer from $3d^8 4s^2$ to $3d^{10} 4s^0$ configuration, so all the *d*-orbitals are paired where higher d occupation led to the absence of magnetic moment of Ni adatom.

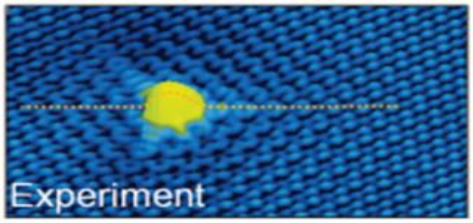


Figure 2: STM topography of a single H atom on graphene [5]
https://inano.medarbejdere.au.dk/news-and-events/events/show/artikel/specialized-inano-lecture-atomic-scale-control-of-graphene-magnetism-using-hydrogen-atoms/

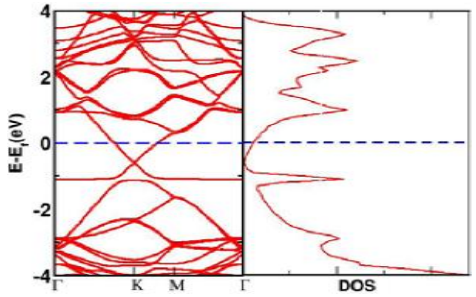


Figure 3: Asymmetric band structure & DOS of Mn adsorbed graphene monolayer [6]
https://www.nepjol.info/index.php/JNPhysSoc/article/view/14439

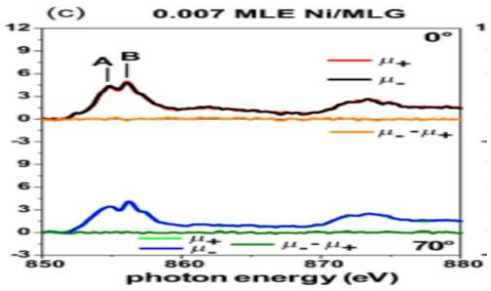


Figure 4: XAS and XMCD spectra obtained for spin up (μ_+) and spin down (μ_-) indicating non-magnetism($\mu_- - \mu_+ \approx 0$)in Ni adsorbed graphene monolayer obtained at 0° and 70° gracing incident angles [10]
https://doi.org/10.1103/PhysRevLett.110.136804

Furthermore, Table 1 shows the magnetic moment variation by adsorbing rare-earth atoms at the H, T and B sites on graphene [18]. By observing the data in the table, it can conclude that the magnetic moment increases gradually when the atomic number, hence the unpaired electrons in the 5*d*, 4*f* or 4*f* orbitals, of the adsorbing the lanthanide series elements increase respectively (Figure 5). Here the sequence of unpaired electrons is 2, 4, 6, and 7 respectively for Ce, Nd, Sm and Eu.

Even though experimental techniques had succeeded in hydrogenated graphene, further developments in efficiency, stability, environmental suitability, and cost-effectiveness are necessary to study. Furthermore, adatom adsorbed graphene has the possibility of being utilised in biological sensors, energy conversion, energy storage devices, spintronics and as well as catalysts which are dependent on the adatom's nature [3].

lanthanide series

Adatom	**Electronic configuration of the adatom**	**Site**	**Magnetic moment(μ_B)**
Ce	[Xe] $4f^1$ $5d^1$ $6s^2$	T	2.903
		H	2.957
		B	4.744
Nd	[Xe] $4f^4$ $6s^2$	T	4.936
		H	4.960
		B	7.024
Sm	[Xe] $4f^6$ $6s^2$	T	7.042
		H	7.034
		B	-8.017
Eu	[Xe] $4f^7$ $6s^2$	T	8.019
		H	8.033
		B	8.036

Table 1: Magnetic moment of adsorption of lanthanide series adatom on graphene monolayer on different sites. [18]

Figure 5: The optimized atomic structure of Eu adatom adsorbed on the H site. Green-Eu adatom, brown- C atoms [18]
https://doi.org/10.4028/www.scientific.net/KEM.645-646.40

2.2 Vacancy Creation

During the production procedure of graphene, lattice defects are formed which led to variations in the lattice structure and eventually induces the magnetism on graphene monolayer. It is highlighted that the magnetic moment is subtle to defect concentration, and vacancy separation [19, 20]. A mono vacancy in the lattice as shown in Figure 6, makes three σ

dangling bonds resulting from the absence of the π electron. As a result, the magnetic moments of the three C atoms with dangling bonds around a vacancy are summed up and produce the local magnetic moment [8]. Figure 7 shows how the spin symmetry (spin up & spin down) has broken in the density of states (DOS) graph due to the mono vacancy created in a 4× 4 superlattice system (Figure 7).

Many DFT calculations on single vacancy monolayer graphene have reported the existence of magnetic moment in the range of 0.69-1.53 μ_B [19,21]. In order to explore the effectiveness of the nature of the vacancies' in the magnetization of graphene STM images of graphene monolayer were been used [22]. Further, this experimental study has clarified the presence of magnetism in vacancy-defected monolayer graphene [22]. They have stated, *"vacancies in single layer graphene lead to magnetic couplings of arbitrary sign"* [22].

Figure 6: Single vacancy monolayer graphene (4*4 superlattice)

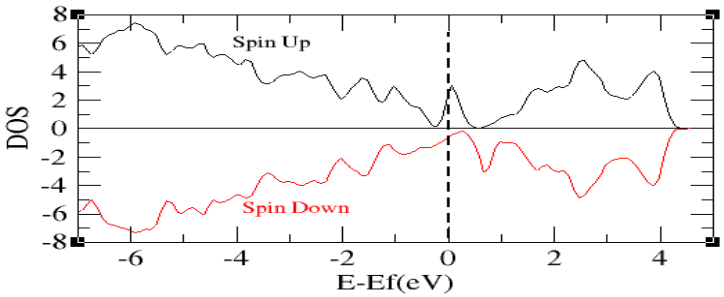
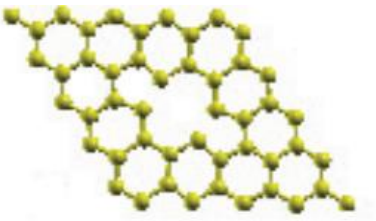


Figure 7: DOS of single vacancy monolayer graphene (4*4 superlattice)

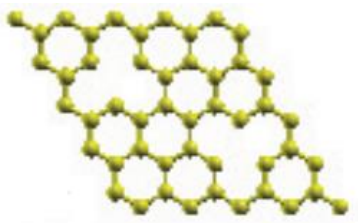
Not only single vacancy but also multi vacancies will trigger the magnetic properties of graphene monolayer [23]. Studies have emphasized that in the vacancy creation method, magnetic properties are mainly affected by vacancy defect concentration and its configuration [23]. For example, experimental and theoretical studies on divacancy had shown a dangling bond-free distorted structure which has been identified as a nonmagnetic system [24, 25]. However, some theoretical studies have shown the existence of nonmagnetic properties in the adjacent divacancy systems while the random divacancy systems exhibit a magnetic moment of 2.00 μ_B. Such two systems are shown in Figure 8 and Figure [9, 26]. Additionally, Dai et al stated*," For the vacancy configurations in which an odd number of C atoms are missing, at least one dangling bond persists, and thus the local moments are present showing ferromagnetic coupling between C atoms at the vacancy defect*" [23]. Therefore, according to

the above observations, it can be concluded that random divacancy is much better than the monovacancy system when concerning magnetic moments. Furthermore, Table 2 shows how the vacancy concentration affects the magnetic moment [27]. According to the table information, magnetic moment decreases when the vacancy concentration increases which implies that the magnetism of this technique strongly depends on the vacancy defect concentration. As of these results, magnetization optimized around 1% to 1.4% of defect concentration. The possible reasoning for the reduction of the magnetic moment beyond the 1.4 % defect concentration may be due to the breaking of randomness when the vacancy concentration increases.



Magnetic moment = 0.00 μ_B

Figure 8: Adjacent divacancy lattice structure in graphene monolayer [26]
https://doi.org/10.1063/1.4980372



Magnetic moment = 2.00 μ_B

Figure 9: Random divacancy lattice structure in graphene monolayer [26]
https://doi.org/10.1063/1.4980372

Vacancy concentration	12.5%	5.6%	2.0%	1.4%	1.0%
Magnetic moment, μB	1.00	1.55	1.91	2.00	2.00

Table 2: Magnetic moment dependency on vacancy concentration [27]

Studies have shown that magnetic properties can be further improved by adsorbing adatoms around the vacancy defected areas [28]. Experimentally, it has been studied the magnetism of divacancy monolayer graphene adsorbed with Si atoms shows sp^2d hybridization [28]. Furthermore, DFT calculations had carried out on binding metal atoms (Pt, Pd, Au, and Sn) on mono vacancy defected graphene monolayer [29] and the magnetic moment values are shown in Table 3 . By observing the Table 3 it can be shown that Au metal atom around a single vacancy, magnetic properties can be further developed. Additionally, adsorbing three

hydrogen atoms around the vacancy area had shown a magnetic moment of 0.15 μ_B [30] as shown in Figure 10.

Adatom	Magnetic moment μ_B
Pt	0.00
Pd	0.00
Sn	0.00
Au	1.00

Table 3: Magnetic moment of by binding metal atoms on single vacancy area [29]

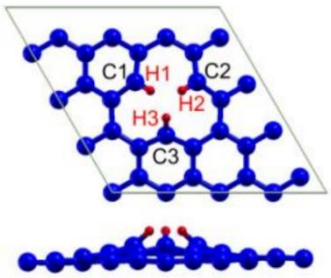


Figure 10: Three H atoms adsorbed around the single vacancy area [30]
https://iopscience.iop.org/article/10.1088/1742-6596/1951/1/012012/meta

2.3 Substitutional Doping

Substitutional doping is done by replacing carbon atoms in the graphene monolayer where electron-accepting or donating heteroatoms facilitates the electronic and magnetic properties changes [3]. Studies have shown that the magnetic properties differ with the size, type, and concentration of dopant adatoms.

Many studies had considered boron (B) as a suitable element for substitutional doping in graphene monolayer due to its similar configuration to the C atom. Experimentally, it has been observed to have the zero bandgap and to have alterations in the electronic structure for B substituted graphene monolayers which had grown using the chemical vapour deposition method [31]. Additionally, the magnetic properties have been observed in them due to unpaired single p_z electron on the neighbouring C atom [31]. Furthermore, the studies have shown the B substituted graphene to have a promising future in supercapacitor technology as well as in Li-ion batteries due to its high specific capacitance and cyclic stability [3].

Furthermore, sulfur (S) substituted graphene monolayer had shown strong ferromagnetism with the magnetization of 5.5 emu g^{-1} at 2 K temperature which had been attributed to the delocalization of graphene [32]. due to the electron-donating by sulfur and as a result, it enhances the electron density in the graphene system. Additionally, according to the DFT calculations by Tuček et al [32], as shown in Table 4, they have shown the variation of magnetic moment with the sulfur concentration which clarified the dependence of magnetic moment on the dopant concentration during the substitution. Figure 11 shows the magnetic moment distribution of 4.2% of sulfur doping and the projected density of state (DOS) graph which

implies the existence of magnetism [32]. Furthermore, looking into Table 5 [33], it shows that the magnetic moment increases when going down the alkaline earth metal group which implies that in this method, the magnetic moment of graphene also depends on the size and the type of the dopant.

Concentration of sulfur	**Magnetic moment (μ_B)**
4.2%	0.5 μB
6.25%	0.2 μB

Table 4: Magnetic moment dependency on substituted S concentration [32]

Substitution Element	**Magnetic moment(μ_B)**
Be	0.00
Mg	1.95
Ca	1.99
Sr	2.00

Table 5: Magnetic moment of alkaline earth metal substitution in graphene monolayer [33]

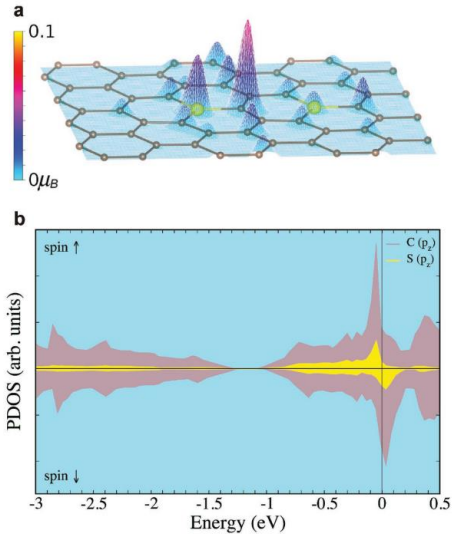


Figure 11: (a)Magnetic moment distribution of 4.2% S doping (yellow-S atoms, brown- C atoms) (b) DOS graph of 4.2% S substitution on graphene monolayer [32]
https://doi.org/10.1002/adma.201600939

2.4 Edge Modifications

Graphene nanoribbons (GNRs) are one of the architectures used in the advancement of spintronics and other magnetism-based devices [34]. GNRs can be attained by cleaving graphene in the armchair and zigzag edge shapes [3] as shown in Figure 12. Theoretical studies have emphasized that the magnetic and electronic properties of GNRs vary with the nature of the atomic edges [3 ,35]. Experimentally, it has been confirmed that there is a finite bandgap in GNRs [36, 37]. The properties of GNRs are different compared to graphene as the symmetrical perfect structure of the graphene monolayer is disturbed by the edge modification of GNRs [35]. A major drawback in GNRs is the difficulty to produce nanoribbons edges with atomic precision and the studied graphene nanoribbons so far are chemically unstable[3,35, 38]. Nevertheless, recently researchers had found a novel experimental method for stabilizing the graphene nanoribbons' edges by substituting Nitrogen atoms and measuring the magnetic properties directly [39]. Theoretically, armchair GNRs are considered to be unfavourable for magnetic ordering [3, 35] while zigzag GNRs are predicted to be having the potential of inducing magnetism and use in spintronics applications [3, 35].

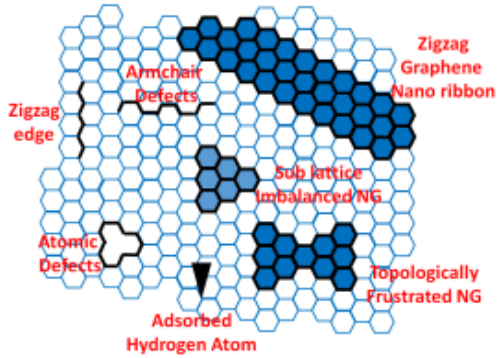


Figure 12: Different types of defects in opening the bandgap in graphene monolayer sheet
https://ieeexplore.ieee.org/document/9388280/authors#authors

Lanfei Xie *et al.*[40] have reported the presence of ferromagnetism at room temperature in partially hydrogenated epitaxial graphene with zigzag edges which had been confirmed by Superconducting Quantum Interference Devices Measurements. According to their suggestions [40], partial hydrogenation forms unpaired electrons which is resulting in the bonding delocalization system and eventually causing ferromagnetism [40].

An experimental study has shown a saturation magnetization of 0.303 emu/g for the GNR with N-doped zigzag edge [34]. Many adatoms and chemical groups have been used to functionalize the graphene edges [2, 34]. Additionally, it is studied and concluded that by functionalizing the one side edge with H, SO_2, and NO_2 atoms, half-metallic zigzag GNRs can be obtained [41]. Wide-range of studies on GNRs have suggested future potential applications in spintronics, energy storing devices, optoelectronics, sensors, high-performance nano-electronics, bioimaging devices etc.[2].

3. DISCUSSION

Even though graphene has many potential novel applications due to its unique properties, zero magnetism can be observed in pristine graphene because of the zero-band gap at the Dirac point and the absence of unpaired electrons. As a result, it restricted the applications on magnetic-based devices such as spintronics. Therefore, many scientists are focused on improving the magnetic properties of graphene so that it can be readily applied to magnetic-based devices. Some possible techniques are reported for inducing magnetism into a graphene monolayer. Out of these techniques, surface doping with adatoms, vacancy creation, substitutional doping and, edge modifications methods have shown favourable outcomes so far.

By adsorbing adatoms, it distorted the lattice configuration and eventually an electric dipole moment and a spin-polarized magnetic moment emerged. The magnetic value of this method solely depends on the nature of the adsorbed adatom. Though the adsorption mechanism had already proven the alteration of magnetism of graphene monolayer, there are many challenges related to this method, especially adsorption of alkaline earth metals is yet to be tested by experimentalists. Even though experimental techniques had succeeded in hydrogenated graphene, further developments in efficiency, stability, environmental suitability, and cost-effectiveness are necessary to study. In addition, further studies in this method are required to manipulate the magnetism where it preserves the intrinsic properties of graphene such as high charge conductivity. Though there is a reliable success in the halogenation of graphene, new challenges are yet to be addressed by experimentalists when considering commercial production.

Unlike the adsorption of the adatom method, the vacancy creation method had gone under many experiments since vacancies are the most common lattice defects seen during its

growth and irradiation. However, the existence of magnetism depends on the no. of vacancies and its configuration where the reasons for these conditions are yet to be studied by researchers. Although there are already many reliable techniques used to create vacancies in graphene monolayer, there are many challenges in commercializing this method and controlling the number of vacancies. Even though theoretically the predicted magnetic moment of a single vacancy graphene monolayer is in the range of 0.69-1.53 μ_B, experimental evidence on room temperature ferromagnetism is still limited. The reasons behind these limitations of room temperature ferromagnetism in graphene monolayer are remained debatable and need further studies. In addition to that, studies have shown that the magnetic moment can be further improved by doping adatoms around the defected area.

Substitutional doping by replacing carbon atoms in the graphene monolayer along with electron-accepting or donating heteroatoms tailored to the electron and magnetic properties. Where the magnetic moment differs with the dopant adatoms' size, type, and concentration. Band gap opening by B and S substitution is dependent on the concentration and room temperature ferromagnetism in this method is yet to be addressed. Though many successful synthesis methods and techniques had developed on substitution methods, large-scale commercialized production is still not yet achieved as it has many challenges. This is because the band gap of this method depends on the concentration and experimentalists have to encounter challenges in controlling the dopants at specific locations in the lattice without affecting the intrinsic properties of graphene.

The edge modification is a reliable method of inducing magnetism in a graphene monolayer where the magnetic properties vary with the nature of the edges. Many theoretical studies had proven the improvement of magnetism in graphene monolayer through this method, but only fewer experiments can be observed in this technique. Although the GNR has been widely developed, functionalizing the edges experimentally is remained difficult to achieve at the atomic scale which is a major drawback of this method.

4. CONCLUSION

Graphene is a promising 2D material with novel applications in nano-electronics, sensors, energy-storing devices, etc. due to its properties like high charge carrier mobility and mechanical properties. However, due to the zero bandgap and non-magnetism, it restricted its wide applications. Hence, this review mainly focuses on four possible magnetism-

inducing methods in graphene monolayers which are surface doping with adatoms, vacancy creation, substitutional doping, and edge modification. In these techniques, the magnetic properties are dependent on the adatom nature, no. of vacancies, vacancy configuration, and the type of edges. The addressed theoretical and experimental evidence support the achievability and the challenges of these methods. One of the challenges is obtaining the magnetism existence in graphene monolayer at room temperature because most of the experimental studies that had been carried out so far had been achieved at low temperatures. Nevertheless, all the mentioned methods have been proven the ability to induce magnetism and have a promising application, especially in spintronics, supercapacitors, batteries, and memory storage devices. Herein, this provides insights to improve the magnetism by altering the electronic structure to induce magnetic moments in graphene monolayers as well as multiple graphene layers.

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