Proceedings of CONF-MSS 2026 Symposium: Mechanical Control and Automation
DOI: 10.54254/2755-2721/2026.BJ31459

Analysis of the Mechanism of Room-Temperature Ferromagnetism in Graphene

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Abstract. In the current world, the demand of memory storage and information transportation materials is gradually increasing, and we should pay more attention to the search for new materials and the development of new applications of existing materials.This study focuses on the pathways to induce ferromagnetism in graphene, as well as the main physical mechanisms that stabilize ferromagnetic order. Although pure graphene typically exhibits diamagnetism due to the electronic characteristics of its carbon atoms, recent research has successfully realized ferromagnetism through a variety of controllable methods. Strategies to achieve ferromagnetism in graphene normally include chemical doping with magnetic atoms and physical method, such as vacancies or edge states. The significance of this study lies in systematically reviewing and elucidating effective approaches to modulate ferromagnetism in graphene and their principles, thereby deepening the understanding of the interplay between magnetism and electronic correlations in low-dimensional materials and opening up potential applications in the fields of spintronics.

Keywords: Two-dimensional magnetic material, Room-temperature ferromagnetism in graphene, Induction method

1. Introduction

The continuous development of information technology is approaching the physical limits of traditional silicon-based electronics, and exploring information processing and storage technologies based on new materials has become a focus of global technological competition.

Two-dimensional materials, such as graphene, have excellent physical properties and are also ideal spin transport media. If stable ferromagnetism at room temperature can be achieved in graphene, it could pave the way for building carbon-based spin logic and storage units, providing a new materials platform for an information technology revolution in the post-Moore era. As a pure carbon material, intrinsic graphene does not possess magnetism. How to induce and stabilize its magnetism has been a hot topic for a long time. Previous studies have mainly followed two approaches: chemical doping and strain induction.

Chemical doping, for example, an early chemical doping method proposed by Sun Q, showed that when hydrogen atoms selectively adsorb on carbon atoms of one lattice in the graphene, the π bonds at those sites are disrupted, causing neighboring non-hydrogenated carbon atoms to generate

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unpaired 2p electrons. These unpaired electrons' magnetic moment can interact through long-range exchange interactions to form stable ferromagnetism [1].

There are also other relevant studies. For example, researchers also found that graphene grown via CVD on copper substrates exhibits strong local strain. This large strain can generate pseudo magnetic fields.It does not exhibit ferromagnetism on a macroscopic level, but still requires attention [2]. In another study in 2025, Wang and colleagues designed and fabricated 'Janus' graphene nanoribbons, with one edge perfectly zigzag and the other modified by periodic topological defects. This design successfully enhanced ferromagnetic ordering along the zigzag edge, theoretically realizing the construction of edge ferromagnetic channels [3].

Through the studies listed before, we have a basic understanding of the ferromagnetic properties in graphene. Although the design of Janus nanoribbons is excellent, the key issues, such as experimental fabrication, stability of magnetism at room temperature, and controllable integration as functional units, remain to be solved.

2. The origin of ferromagnetism in graphene

The ferromagnetism of graphene is caused by the magnetic moments of its electron spins and typically needs controlled symmetry breaking or coupling with external magnetic moments to create ferromagnetism. Its fundamental physical process can be explained by the creation of magnetic moments and their long-range correlations.

Intrinsic magnetism in graphene is also originates from local atom's magnetic moments. In a perfect lattice of graphene, because of the character of its p electrons and symmetry of the lattice, the electron spins are paired, and it is diamagnetic. However, when the periodicity and symmetry of the lattice are disrupted, such as by introducing vacancies, the situation changes fundamentally. Take a single vacancy as an example: the absence of a carbon atom leaves three unsaturated dangling bonds.These bonds undergo reconstruction, with one forming a new ring, leaving a stable localized state. This localized state is close to the Fermi level and manifests as a peak in the electronic density of states. According to Stoner's criterion, when the density of electronic states is sufficiently high and electron correlation is strong enough, the system will spontaneously undergo spin polarization to reduce exchange energy, thereby generating local magnetic moments at defect sites. Theoretical DFT calculations indicate that the edge carbon atoms, due to their unique edge geometry, possess electron states similar to high density, which may lead to edge state spin polarization [4].

The magnetic moments produced by the individual defects are however isolated. In order to obtain macroscopic ferromagnetism, there is need to have a good ferromagnetic coupling between these local magnetic moments. This interaction is usually thought to be mediated by conduction electrons of graphene, through the RKKY interaction. These local magnetic moments polarize the surrounding conduction electron cloud and this spin polarization propagates in an oscillatory manner through space, interacting with other distant local magnetic moments [5]. The ferromagnetic and strength of this interaction depend strongly on the Fermi wave vector and spacing between the magnetic moments. Introducing high concentration and specific spatial distribution of defects makes it theoretically possible to create long-range ferromagnetic order but experimentally, stable and controllable room-temperature ferromagnetism remains very difficult to achieve.

Another more effective approach is to add exogenous magnetic centers, namely atomic doping. Magnetic transition metal atoms such as Fe, Co, and Ni, or light atoms with unpaired p electrons such as H and F can be incorporated into the graphene lattice through chemical adsorption and doping. The mechanism is that magnetic atom centers are indirectly coupled through the polarization

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of the conduction electron cloud, which provides a reliable physical basis for achieving long-range ferromagnetic order.

Overall, the ferromagnetism of graphene is not a fundamental property but rather a result of functional changes in its nonmagnetic ground-state electronic structure, which adds localized magnetic moments by means of defects or doping and creates long-range correlations between magnetic moments through the conductive properties of the electron system. The central scientific requirement to design and prepare practical graphene based spin logic devices is an intimate understanding of this physical picture.

3. Main methods for regulating the room-temperature ferromagnetism of graphene

3.1. Chemical doping

3.1.1. Doping of non-metallic elements

Nitrogen, as one of the most commonly used dopants, can form three types of doping configurations in graphene: pyrrolic N, graphitic N, and pyridinic N. Graphitic nitrogen is located inside the hexagonal rings of graphene, directly replacing a carbon atom and bonding with three surrounding carbon atoms through σ bonds. Pyridinic nitrogen is usually found at the edges or defect regions of graphene, bonding with two carbon atoms in a coordinated manner while introducing an unpaired lone pair of electrons. Pyrrolic nitrogen is typically associated with five-membered ring structures, bonding with one carbon atom in a singly coordinated manner and contributing two π electrons [6].

According to the experimental results of researchers Jiayuan Li et al., pyridine-type graphene was synthesized using the hydrothermal method, and nitrogen doping was carried out by treating graphene oxide with ammonia at low temperatures (≤600°C). It was found that its saturation magnetization rose by 64.1%. This means that chemical nitrogen doping managed to attain stable magnetic characteristics in graphene through the creation of asymmetrically distributed local magnetic moments.

Hydrothermal synthesis of doped graphene is not only a way to synthesize nitrogen-doped graphene. As the study by Gunja et al. indicates, boron-doped graphene produced using hydrothermal method also shows similar magnetism and the saturation magnetization (Ms) increases with the amount of boron. The sample having the maximum concentration of boron achieved a saturation magnetization (Ms) of 0.03 emu/g and coercivity (Hc) of 73.9 Oe at room temperature. This proves that boron-doped graphene oxide has considerable ferromagnetism at room temperature [7].

The benefit of the hydrothermal method is its ability to precisely control morphology and structure of complex materials, as well as its green synthesis properties, which make it ideal for innovative functional material research, especially in nanomaterials and low-dimensional materials. However, its safety, complex post-processing, and industrial adaptability require special attention.

3.1.2. Transition metal element doping

Magnetism can also be increased by doping with transition metals. As an example, room-temperature ferromagnetism up to 400K can be achieved through embedding Co-N4 coordination groups into graphene. In particular, isolated cobalt atoms coordinate with nitrogen atoms to form a planar square structure, leading to a pronounced ferromagnetic hysteresis loop and strong magnetization. DFT calculations show that the d electrons of cobalt hybridize with the electrons of

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nitrogen and carbon atoms, increasing long-range magnetic coupling mediated by conduction electrons. DFT simulation results also indicate that the Co-N4 structure is responsible for the observed room-temperature ferromagnetism [8].

3.1.3. Oxygen-containing organic functional group modification

The presence of oxygen-containing functional groups can be used to induce ferromagnetism in graphene, which is simply the introduction of defects. Oxygen-containing functional groups that are covalently bonded to the basal plane of the graphene interfere with the sp^2 hybridization of carbon atoms and make the bonded carbon atoms shift to sp 3 hybridization. This reaction highly localizes the electrons on the pi-electron network of the graphene, producing unpaired electrons or localized states on the adsorption sites of the functional group or adjacent carbon atoms. These spins of unpaired electrons are the source of the induced local magnetic moments. Zhang, D et al. showed that by accurately controlling the reduction process of graphene oxide, the spatial distribution of residual oxygen-containing functional groups such as epoxy and hydroxyl groups on the carbon basal plane can be effectively regulated [9]. Experiments confirmed that when functional groups cluster to form specific aggregate structures rather than being highly dispersed during the reduction process, significant and stable room-temperature ferromagnetism can be induced in the graphene lattice.

3.2. Stress engineering

Stress engineering is a cutting-edge technology for regulating the electronic properties of graphene, and one of its goals is to induce pseudo magnetism without the need for traditional magnetic elements. Although this magnetic field does not exhibit ferromagnetism on a macroscopic scale, its generation is also noteworthy.The core physical principle of this method is that applying non-uniform strain to a single layer of graphene will change the bond length and Angle between carbon atoms, thereby introducing an equivalent gauge field into the low-energy effective theory. This gauge field is equivalent to a powerful local magnetic field, that is, a pseudo magnetic field, for the unique Dirac electrons in graphene. This field can quantize electronic states into Landau energy levels just like a real magnetic field, resulting in characteristic peaks in the local electronic state density and possibly triggering magnetic correlation phenomena such as the quantum Hall effect.

Among them, N.C. Yeh deposited graphene on copper substrates by chemical vapor deposition [10], and studied the effect of nanoscale strain on electronic properties at low temperatures (77 K and 300 K) with STM. It has been discovered that the thermal mismatch between the expansion of graphene and the contraction of copper during the cooling process creates a strong non-uniform strain and corrugated structure on a copper substrate, which in turn causes a pseudo magnetic field as high as 50T and a charging effect.

On the basis of this, strain engineered graphene can be designed to mimic pseudo magnetic fields by regulating strain using substrate textures and generate transport properties with a large on-off ratio that has potential applications.Although there is currently no research indicating that this specific magnetic field can exhibit macroscopic ferromagnetism, there is potential for regulation in the future.

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3.3. Defect regulation

The study of edge states has attracted significant recent interest and the corresponding mechanisms are also complex. The magnetic moments generated in graphene are due to the local magnetic moments formed by unpaired electrons of carbon atoms at the zigzag edges. In traditional symmetric zigzag nanoribbons, the magnetic moments at the two edges show overall antiferromagnetism because of antiferromagnetic coupling. Janus graphene nanoribbons (JGNRs) asymmetrically introduce periodic topological defects along one zigzag edge in their design. These defects, made up of benzene rings, quench the intrinsic magnetic moments on that edge so that the magnetic moments are entirely localized on the other, intact zigzag edge.

The study by Shaotang Song, which is an integration of a specifically designed surface synthetic chemistry and high-resolution scanning probe methods, was the first to experimentally succeed in building and verifying that Janus graphene nanoribbons are able to have stable intrinsic ferromagnetic order. The experimental aspect is based on a new Z-shaped precursor molecule, which, by controlled thermal annealing on Au(111) single crystal surface can be successfully converted into two JGNR structures with widths parameterized as (4,2) and (5,2). Atomic-scale structural characterization by STM and AFM unambiguously showed asymmetric topological structure: one edge had a topological defect array (TDZ edge) made of benzene units with regular spacing, whereas the other edge was intact, undisturbed zigzag edge (ZZ edge). This directly proves the experimental implementation of the theoretically developed topological edge engineering.

To investigate the electronic and magnetic states, the research team further used STS to measure the local density of electronic states. Characteristic peaks near the Fermi level were found directly above the intact ZZ edge, which means these states are strongly localized at this edge. In sharp contrast, these characteristic peaks completely disappeared at the corresponding positions of the modified TDZ defect edge. These STS results give direct experimental evidence that confirms the theoretically predicted phenomenon of spin-polarized edge states being localized on a single edge.

In combination with DFT calculations, these localized electronic states were definitely assigned to the spin-polarized states created by local magnetic moments of the ZZ edge carbon atoms. Further calculations have also shown that the ground-state coupling of these magnetic moments, isolated on one side, is ferromagnetic, and the band gaps of spin-polarized are calculated as 0.21 eV and 0.19 eV, respectively, meaning that this ferromagnetic state is stable enough at room temperature. All the chain of experimental evidence, including the determination of atomic structures, mapping the spatial distribution of electronic states, and theoretical ground-state calculations, is highly interrelated and self-consistent, which ultimately proves that Janus structures with single-sided topological defect modification can quench magnetism on one edge but induce and stabilize ferromagnetic order on the other intact zigzag edge.

4. Conclusion

There are a number of ways to achieve room-temperature ferromagnetism in graphene, and they can be categorized into two exploratory directions: first, by chemical doping, adding local magnetic moments and coupling them into ferromagnetic order using long-range exchange interactions. Second, by designing structures that break symmetry, such as by modulating edge state magnetic order. These studies provide important theoretical mechanisms and experimental ideas for inducing and stabilizing room temperature ferromagnetism in graphene.

In the future, studies ought to be directed towards building macroscopic graphene ferromagnetic systems that are stable at room temperature. On the one hand, it is necessary to develop scalable and

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accurate atomic-level fabrication techniques to attain controllable magnetic order; on the other hand, new applications, transport, and detection methods based on the ferromagnetic properties of graphene should also be explored to further develop all-carbon spin logic and storage units into reality.

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