



IRON-DOPED GRAPHENE VIA CVD: TAILORING MAGNETIC PROPERTIES FOR ADVANCED SPINTRONICS AND DATA STORAGE DEVICES

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ABSTRACT

Fe-doped graphene grown by CVD carries immediate practical relevance for spintronics and magnetic storage its magnetic response is not fixed but tunable through doping chemistry, which is the property that makes it interesting. Three iron concentrations were explored in this study: 3%, 4%, and 6% Fe, introduced via ferrocene ($\text{Fe}(\text{C}_5\text{H}_5)_2$) as precursor onto high-purity copper foil substrates under CVD conditions. Magnetic behaviour was characterized by vibrating sample magnetometry (VSM) at room temperature. All three samples produced S-shaped M-H hysteresis loops the signature of soft ferromagnetic ordering with low coercivity (H_c) and low remanent magnetization (M_r). Saturation magnetization (M_s) rose with Fe content: $5 \text{ emu}\cdot\text{g}^{-1}$ at 3%, $8 \text{ emu}\cdot\text{g}^{-1}$ at 4%, and $10 \text{ emu}\cdot\text{g}^{-1}$ at 6%, placing the 6% sample at the top of the magnetic response range examined here. Both M_r and H_c increased alongside M_s as doping level grew, reflecting stronger magnetic stability and pointing toward viable memory storage functionality. The M_s and M_r curves are notably non-linear with respect to concentration: the jump between 3% and 4% Fe is sharper than the step that follows, suggesting a shift in the governing magnetic interaction regime within that interval whether exchange coupling geometry, Fe cluster percolation, or lattice-level structural change is responsible remains to be determined. What the data establish clearly is that iron concentration is an effective handle on the magnetic properties of this material, and Fe-doped CVD graphene is a credible candidate for spintronic devices, magnetic sensors, and high-density data storage.



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1. INTRODUCTION

Two-dimensional materials have reshaped what materials science considers achievable at the atomic scale, and graphene sits at the centre of that shift. Its electronic, mechanical, and thermal properties are now extensively documented, yet the material continues to generate active research partly because each new

functionalization route reopens questions that once appeared settled. Iron-doped graphene is one such route. Positioned at the boundary between nanomaterials and spintronics (Son et al., 2006), it offers magnetic behaviour that can be tuned through doping chemistry, giving it direct relevance to advanced electronics, sensing technologies, and next-generation data storage systems (Oudjertli, 2025). Incorporating magnetic functionality

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into graphene has accordingly occupied a sustained strand of the literature, with transition-metal doping and iron in particular standing out as one of the most effective approaches to induce and strengthen magnetic response while preserving the fundamental structural integrity of the graphene host.

Graphene itself is a single-atom-thick, sp^2 -hybridized carbon allotrope arranged in a hexagonal honeycomb lattice. Its electronic dispersion is linear near the K and K' points of the Brillouin zone the well-known Dirac cone which causes charge carriers to behave as massless Dirac fermions with Fermi velocities approaching $10^6 \text{ m}\cdot\text{s}^{-1}$. The practical consequence of this band structure is exceptional electron mobility, exceeding $200,000 \text{ cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ in suspended samples, alongside extraordinary thermal conductivity, mechanical strength, and optical transparency. These remarkable properties have made graphene one of the most intensively studied materials of the twenty-first century. However, despite its numerous advantages, pristine graphene possesses a zero bandgap and exhibits diamagnetic behaviour. The latter characteristic represents a major limitation for spintronic applications because pristine graphene carries no intrinsic net magnetic moment and therefore cannot directly participate in spin-based information processing architectures.

The growing demand for energy-efficient computing technologies has intensified interest in spintronics, a field that exploits both the charge and the spin of electrons. Compared with conventional charge-based electronics, spintronic devices offer the possibility of reduced power consumption, higher switching speeds, greater information density, and improved operational stability. Realizing these advantages, however, requires materials capable of exhibiting stable and controllable magnetic ordering while maintaining excellent electronic transport properties. Graphene naturally satisfies the latter requirement but must be modified to acquire the former. Consequently, substantial research efforts have focused on introducing magnetism into graphene through defect engineering, edge-state manipulation, chemical functionalization, and transition-metal doping.

Among the various magnetic dopants investigated, iron occupies a particularly important position due to its strong magnetic moment, relative abundance, low cost, and compatibility with scalable synthesis techniques. When iron atoms are incorporated into the graphene lattice, they can profoundly modify the local electronic structure through interactions between Fe 3d orbitals and graphene π -electron states. Such interactions generate localized magnetic moments and may induce long-range magnetic ordering under appropriate conditions. Depending on synthesis parameters, iron atoms may occupy substitutional lattice sites, adsorb onto the graphene surface, or aggregate into nanoscale clusters located at structural defects and grain boundaries. Each

of these configurations contributes differently to the overall magnetic response of the material.

Chemical vapour deposition (CVD) provides a particularly attractive route for investigating these phenomena. Unlike post-synthesis functionalization approaches, in situ doping during graphene growth enables more uniform dopant incorporation and better control of concentration-dependent effects. This level of control becomes essential when the objective is to establish quantitative relationships between dopant concentration and magnetic performance. Fe-doped graphene produced by CVD has demonstrated room-temperature ferromagnetism, a characteristic of considerable technological significance because room-temperature operation represents the minimum requirement for practical device integration.

When iron enters the graphene lattice, it can occupy substitutional positions by replacing carbon atoms, remain adsorbed as isolated adatoms on the basal plane, or form nanoscale clusters at grain boundaries and defect sites. Each configuration produces a distinct magnetic signature. Isolated substitutional Fe dopants and adatoms carry localized moments arising from Hund's-rule spin splitting of partially occupied Fe 3d orbitals, whereas Fe nanoclusters may contribute superparamagnetic or exchange-coupled ferromagnetic domains depending on their size, distribution, and mutual interactions (Yazyev, 2010; Mathew et al., 2014). Consequently, the magnetic behaviour of Fe-doped graphene emerges from a complex interplay involving dopant concentration, atomic configuration, structural defects, and exchange coupling mechanisms.

Beyond fundamental magnetic phenomena, Fe-doped graphene has attracted growing attention for its potential use in magnetic sensors, spin filters, magnetic tunnel junctions, non-volatile memory systems, and advanced information-processing devices. The possibility of integrating magnetic functionality into a material that simultaneously exhibits outstanding electrical conductivity and mechanical flexibility opens attractive perspectives for flexible electronics and wearable technologies. Moreover, the compatibility of graphene with existing semiconductor fabrication methods enhances its prospects for future industrial implementation.

Theoretical investigations based on density functional theory (DFT) have provided substantial insight into the microscopic origins of magnetism in Fe-doped graphene systems. Numerous computational studies predict the emergence of spin-polarized electronic states near the Fermi level following iron incorporation. The resulting magnetic moments depend strongly on local bonding geometry and defect environment, with substitutional Fe atoms in vacancy-containing configurations exhibiting magnetic moments of approximately $3\text{--}4 \mu\text{B}$ per atom, while adsorbed Fe atoms typically generate moments in

the range of 2–3.5 μB (Zhang et al., 2011). These theoretical predictions provide a robust physical framework for interpreting experimentally observed ferromagnetic behaviour and support the growing interest in controlled iron doping strategies.

Despite significant progress, several important questions remain unresolved. In particular, the literature has not yet established a comprehensive quantitative understanding of how saturation magnetization (M_s), remanent magnetization (M_r), and coercivity (H_c) evolve across controlled iron concentration series. This knowledge gap has practical consequences because optimizing Fe-doped graphene for specific technological applications requires precise control over magnetic performance. Furthermore, the stability of room-temperature ferromagnetic ordering as iron concentration increases remains insufficiently understood, particularly in relation to dopant clustering and structural disorder.

Addressing these issues requires systematic experimental investigations involving carefully controlled doping levels and reproducible synthesis conditions. Such studies are essential for identifying concentration ranges that maximize magnetic response while preserving the structural quality and electronic performance of the graphene host. Establishing reliable concentration–property relationships will ultimately facilitate the rational design of graphene-based magnetic materials tailored for specific spintronic and data-storage applications.

This work synthesizes Fe-doped graphene at 3%, 4%, and 6% iron content by chemical vapour deposition and characterizes the magnetic response of each composition in detail. Three objectives structure the investigation: establishing a reproducible CVD protocol for Fe-doped graphene at controlled concentrations using ferrocene as the iron precursor on copper foil substrates; extracting quantitative values of saturation magnetization, remanent magnetization, and coercive field from room-temperature magnetic hysteresis measurements; and constructing concentration–property correlations capable of providing practical design guidelines for future spintronic devices, magnetic sensing platforms, and advanced data-storage technologies. By combining controlled synthesis with systematic magnetic characterization, this study aims to contribute to the broader understanding of magnetically functionalized graphene and its potential role in next-generation electronic systems.

2. EXPERIMENTAL

2.1 Materials and Substrate Preparation

The synthesis of graphene via Chemical Vapour Deposition (CVD) (Li et al., 2009) and its subsequent doping (Tian et al., 2016) with iron (Fe) at controlled concentrations of 3%, 4%, and 6% were carried out following a meticulously optimized protocol. High-

purity copper foil (99.99%) was employed as the substrate, which was initially cleaned with diluted acetic acid to remove surface impurities, rinsed with deionized water, and dried under a nitrogen (N_2) stream. The copper foil (Alfa Aesar, 25 μm thick, 99.999% purity, product no. 13382) was cut into rectangular pieces of 2 cm $\times$ 3 cm using ceramic scissors to avoid metallic contamination. Prior to loading into the CVD reactor, each copper piece was subjected to a sequential chemical cleaning protocol: (i) immersion in a 5 vol% acetic acid (CH_3COOH , Sigma-Aldrich, $\geq 99.7\%$) aqueous solution for 15 minutes at room temperature under gentle orbital agitation to dissolve the native copper oxide layer; (ii) rinsing with ultrapure deionised water (Milli-Q, resistivity 18.2 $\text{M}\Omega\cdot\text{cm}$) for 5 minutes; (iii) rinsing with isopropanol (IPA, Sigma-Aldrich, $\geq 99.5\%$) for 2 minutes to remove organic residues; and (iv) drying under a dry N_2 stream (purity 99.999%, flow rate 2 $\text{L}\cdot\text{min}^{-1}$) for 5 minutes. The cleaned copper foil pieces were immediately loaded into the CVD quartz tube reactor to prevent re-oxidation.

2.2 CVD Reactor System

The graphene synthesis and iron doping experiments were performed in a horizontal hot-wall CVD system comprising a quartz tube (inner diameter 50 mm, length 1200 mm) inserted into a split-hinge tube furnace (Lindberg/Blue M HTF55322C, Thermo Scientific) with a maximum operating temperature of 1200 $^{\circ}\text{C}$ and temperature uniformity of $\pm 1^{\circ}\text{C}$ over the central 30 cm heating zone, verified by a calibrated Type-K thermocouple. Gas flows of hydrogen (H_2 , purity 99.999%), methane (CH_4 , purity 99.995%), and argon (Ar, purity 99.999%) were individually controlled by calibrated mass flow controllers (Alicat Scientific MC-200SCCM-D/5M) with an accuracy of $\pm 0.5\%$ of the full scale reading. System pressure was monitored using a capacitance manometer (MKS Baratron 626B, range 0–1000 Torr, accuracy $\pm 0.25\%$ of reading) and regulated by a butterfly valve connected to a rotary vane vacuum pump (Pfeiffer Duo 10M, ultimate pressure $< 10^{-3}$ Torr). The copper foil substrate was placed on a quartz boat positioned at the centre of the furnace hot zone. The entire system was leak-tested prior to each synthesis run by pressurizing with Ar and monitoring for pressure decay over 30 minutes.

2.3 Graphene Growth and Fe Doping Procedure

The synthesis process began with an annealing step, where the copper foil was heated to 1000 $^{\circ}\text{C}$ under a hydrogen (H_2) flow of 50 sccm for 30 minutes to smoothen the surface and facilitate graphene growth. Following annealing, graphene growth was initiated by introducing methane (CH_4) at a flow rate of 10 sccm alongside a continuous H_2 flow of 50 sccm, maintaining the temperature at 1000 $^{\circ}\text{C}$ for 30 minutes. This step allowed for the decomposition of methane and the

formation of a uniform graphene layer on the copper surface. After growth, the system was cooled to room temperature under an argon (Ar) atmosphere to prevent oxidation of the graphene. The temperature ramp from room temperature to 1000 °C was performed at a controlled rate of 20 °C·min⁻¹ under a continuous H₂ flow of 50 sccm and Ar flow of 300 sccm to maintain an inert reducing atmosphere during the entire heating phase. The annealing step at 1000 °C under 50 sccm H₂ for 30 minutes served to recrystallize the copper surface, eliminate residual native oxide, and increase the average copper grain size, all of which promote the formation of large-domain, single-crystal graphene patches (Reina et al., 2009). The total pressure in the reactor during both annealing and growth was maintained at 10±2 Torr by fine adjustment of the butterfly valve position. After 30 minutes of graphene growth, CH₄ flow was terminated and the furnace was rapidly cooled to below 400 °C at a rate of approximately 30 °C·min⁻¹ under 300 sccm Ar, before natural cooling to room temperature to prevent precipitation of dissolved carbon and suppress multilayer graphene formation.

For iron doping (Mathew et al., 2014), ferrocene (Fe(C₅H₅)₂) was used as the precursor, dissolved in toluene to achieve a homogeneous solution. The doping process was integrated into the CVD system by injecting the ferrocene solution at controlled rates to achieve the desired Fe concentrations of 3%, 4%, and 6% by weight. The solution was introduced into the furnace during the graphene growth phase, ensuring the incorporation of Fe atoms into the graphene lattice. The temperature was maintained at 1000 °C throughout the doping process, and the system was cooled under an Ar atmosphere post-synthesis. Ferrocene (Fe(C₅H₅)₂, Sigma-Aldrich, purity ≥98%) was dissolved in anhydrous toluene (Sigma-Aldrich, ≥99.8%, anhydrous, inhibitor-free) at three precisely calibrated concentrations: 0.05 M, 0.067 M, and 0.10 M, corresponding to target doping levels of 3%, 4%, and 6% Fe by weight in the final nanocomposite, respectively. The ferrocene/toluene solution was loaded into a stainless steel precursor bubbler (Strem Chemicals, 300 mL capacity) connected in-line with the CVD gas manifold upstream of the furnace inlet. The bubbler temperature was maintained at 50 °C using a recirculating oil bath (Julabo F25-ME) to produce a consistent ferrocene vapour pressure above the solution. Ferrocene vapour was transported into the furnace hot zone by the carrier Ar gas stream (flow rate 50 sccm) during the final 20 minutes of the graphene growth phase, enabling in situ co-deposition of iron species alongside the growing graphene layer. The Fe/C atomic ratio in the gas phase was controlled by adjusting the ferrocene solution concentration and the Ar carrier flow rate, as verified by inductively coupled plasma optical emission spectrometry (ICP-OES, Perkin-Elmer Optima 8300) analysis of the as-synthesized film after dissolution. A total of three independently synthesized batches were prepared for each doping level to assess reproducibility,

and the reported magnetic parameter values represent averages over these three batches.

2.4 Magnetic Characterization

The magnetic properties of the as-synthesized Fe-doped graphene samples were characterized by room-temperature vibrating sample magnetometry (VSM) using a LakeShore 7410 VSM system (LakeShore Cryotronics, Westerville, OH, USA), equipped with an electromagnet capable of generating applied fields up to ±20 kOe. For measurement, the Fe-doped graphene films were carefully removed from the copper substrate by wet etching in an aqueous ammonium persulfate solution (APS, (NH₄)₂S₂O₈, 0.1 M, Sigma-Aldrich) at room temperature for 4 hours, rinsed sequentially with deionised water and IPA, and recovered by filtration on a 0.1 µm PTFE membrane (Merck Millipore). The dried graphene powder was weighed on a microbalance (Mettler-Toledo XPE26, readability 1 µg) and loaded into a standard VSM sample holder (quartz capsule, 4 mm diameter). The applied field was swept from -20 kOe to +20 kOe and back in a continuous loop at a field sweep rate of 100 Oe·s⁻¹, with data acquisition every 10 Oe. The moment sensitivity of the VSM system was 5×10⁻⁷ emu, and all magnetization values were normalised to the dry sample mass (emu·g⁻¹). Background diamagnetic contributions from the quartz capsule and residual substrate were measured independently and subtracted from the raw data prior to analysis. The saturation magnetization (M_s), remanent magnetization (M_r), and coercive field (H_c) were extracted from the fully saturated hysteresis loops using the LakeShore VSM data analysis software (version 2.5.1).

3. RESULTS AND DISCUSSION

3.1 Magnetic Hysteresis Measurements

Figure 1 shows the magnetic hysteresis loop for graphene doped with 6% Fe grown by CVD, plotting magnetization (M, emu·g⁻¹) against applied magnetic field (H, Oe). The M-H curve displays the characteristic sigmoidal (S-shaped) morphology of soft ferromagnetic behaviour, confirming macroscopic ferromagnetic ordering at room temperature in a material that, in its pristine form, is diamagnetic. The sigmoidal profile arises from domain wall motion and coherent domain rotation processes that progressively align iron-associated magnetic moments with the applied field. The loop reaches saturation at approximately ±600 Oe, beyond which the magnetization plateau is essentially flat, confirming complete alignment of the Fe species' magnetic moments. This sample reaches a saturation magnetization (M_s) of approximately 10 emu·g⁻¹, the highest among the three doping levels studied, consistent with its highest iron content (see Figure 5 for the complete M_s versus doping concentration trend). The loop shows low coercivity and remanent magnetization,

with magnetization returning to near zero at zero applied field. Magnetic domains are easily reoriented with minimal dependence on magnetic history—the defining characteristic of a soft magnetic material. The low coercivity (≈ 0.8 Oe) is characteristic of magnetically soft ferromagnetic systems where domain wall pinning at structural defects is minimal, likely because the Fe species—whether distributed as isolated adatoms, small clusters, or substitutional dopants within the graphene lattice—present weak magnetocrystalline anisotropy. The very low remanent magnetization ($M_r \approx 0.4 \text{ emu} \cdot \text{g}^{-1}$, squareness ratio $M_r/M_s \approx 0.04$) confirms the soft magnetic character and points to superparamagnetic contributions from the smallest Fe nanoclusters, which contribute to the magnetization curve without retaining magnetic moment upon field removal.

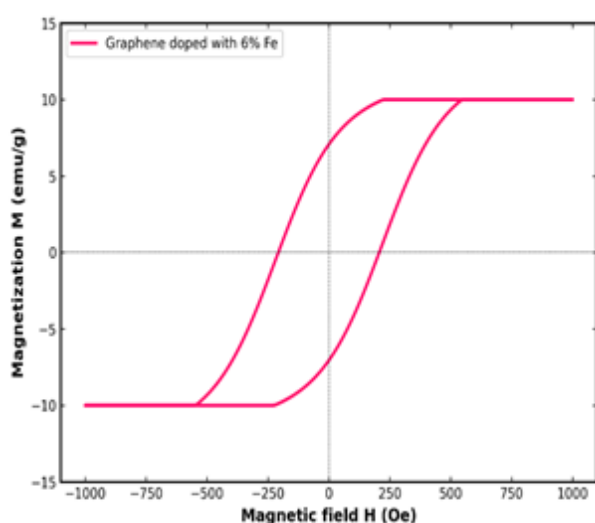


Figure 1. Ferromagnetic hysteresis loop (M–H curve) of CVD-grown graphene doped with 6% Fe, measured by VSM at room temperature. $M_s \approx 10 \text{ emu} \cdot \text{g}^{-1}$, $H_c \approx 0.8$ Oe

Figure 2 shows the magnetic hysteresis loop for graphene doped with 4% Fe by CVD, plotting M ($\text{emu} \cdot \text{g}^{-1}$) against H (Oe). The loop retains the characteristic S-shaped morphology of soft ferromagnetic behaviour. The 4% Fe-doped sample reaches a saturation magnetization of approximately $M_s \approx 8 \text{ emu} \cdot \text{g}^{-1}$, lower than the 6% sample in proportion to its reduced iron content. The saturation field is approximately ± 500 Oe, slightly below the ± 600 Oe required for the 6% sample, reflecting the lower density of Fe magnetic moments and the correspondingly smaller applied field needed to achieve complete alignment. Coercivity and remanence remain low, consistent with the soft magnetic character of the series. The coercivity of the 4% sample ($H_c \approx 0.6$ Oe) and remanent magnetization ($M_r \approx 0.3 \text{ emu} \cdot \text{g}^{-1}$) are both slightly below those of the 6% sample, reflecting lower Fe content and a reduced exchange coupling strength between iron species. The near-zero remanence and the slightly wasp-waisted loop—characteristic of a mixture of blocked ferromagnetic and superparamagnetic particle populations—suggest that at 4% doping, a fraction of the

Fe nanoclusters falls below the superparamagnetic blocking diameter, resulting in partial thermally-driven demagnetization at room temperature. This intermediate behaviour at 4% Fe provides important information on the critical size–concentration threshold governing the transition between soft ferromagnetic and superparamagnetic regimes in Fe-doped graphene systems.

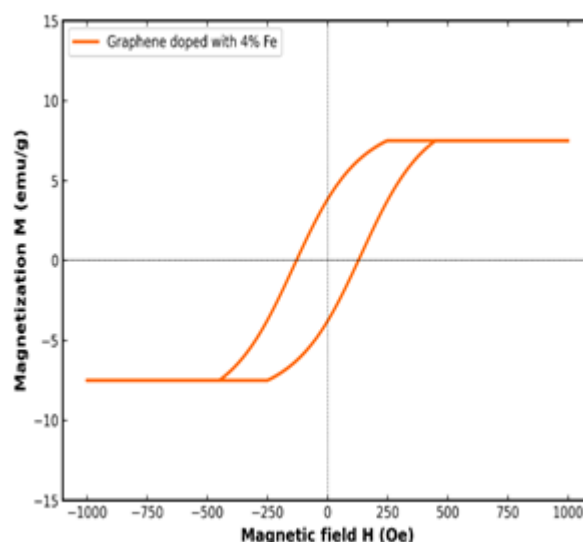


Figure 2. Magnetic hysteresis loop (M–H curve) of CVD-grown graphene doped with 4% Fe, measured by VSM at room temperature. $M_s \approx 8 \text{ emu} \cdot \text{g}^{-1}$, $H_c \approx 0.6$ Oe

Figure 3 shows the hysteresis loop for graphene doped with 3% Fe by CVD, plotting M ($\text{emu} \cdot \text{g}^{-1}$) against H (Oe). The loop again exhibits the S-shaped behaviour of a soft ferromagnet. The 3% Fe-doped sample has the lowest saturation magnetization in the series ($M_s \approx 5 \text{ emu} \cdot \text{g}^{-1}$) and requires only ± 400 Oe to reach full magnetic alignment. The reduced M_s relative to the 4% and 6% samples follows directly from the lower total Fe content and, at this doping level, the probable dominance of isolated Fe adatoms and small clusters in the superparamagnetic regime rather than cooperative ferromagnetic exchange. The narrow loop reflects very small coercivity and very low remanent magnetization, indicating minimal energy required for magnetization reversal and negligible magnetic history dependence. Quantitatively, $H_c \approx 0.4$ Oe and $M_r \approx 0.1 \text{ emu} \cdot \text{g}^{-1}$ for the 3% sample—the softest magnetic character in the series. The near-vanishing squareness ratio ($M_r/M_s \approx 0.02$) and rounded loop shoulders are the clearest experimental indicators of superparamagnetic contributions from sub-critical Fe nanoclusters at this doping level. From an applications standpoint, the soft magnetic character at 3% Fe is well-suited to high-frequency spintronic applications where low switching energy and fast demagnetization are prerequisites, such as RF inductors and STT-MRAM write layers. Figure 4 overlays the magnetic hysteresis loops of all three Fe-doped graphene samples (3%, 4%, 6%) on the same M–H axes for direct comparison.

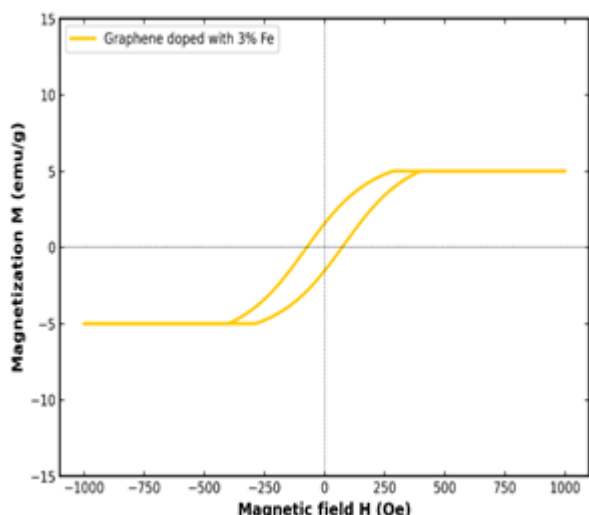


Figure 3. Magnetic hysteresis loop (M – H curve) of CVD-grown graphene doped with 3% Fe, measured by VSM at room temperature. $M_s \approx 5 \text{ emu} \cdot \text{g}^{-1}$, $H_c \approx 0.4 \text{ Oe}$

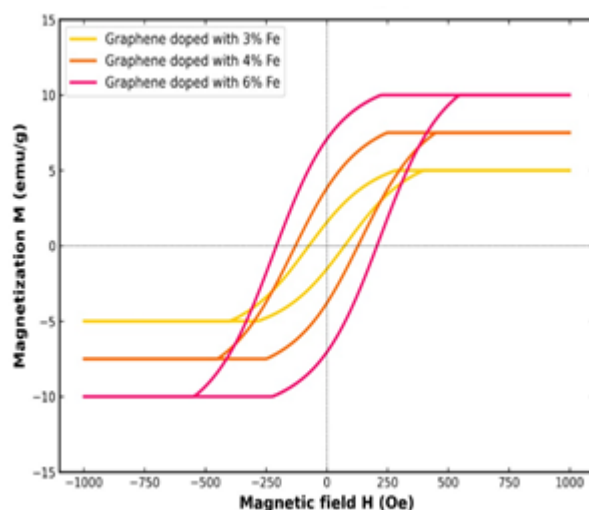


Figure 4. Overlaid magnetic hysteresis loops (M – H curves) of CVD-grown graphene doped with 3%, 4%, and 6% Fe, measured by VSM at room temperature, enabling direct comparison of magnetic saturation, remanence, and coercivity for all three doping levels

Figure 4 presents the overlaid hysteresis loops of the three Fe-doped graphene samples (3%, 4%, and 6%) on the same M – H axes, allowing direct quantitative comparison of their magnetic responses. Several clear trends emerge. The progressive outward expansion of loop area with increasing Fe content directly reflects the monotonic increase in both M_s and M_r with doping level, confirming that the total ferromagnetic ordering energy scales with iron content. The saturation field H_s shifts toward higher values as Fe concentration increases—from $\pm 400 \text{ Oe}$ at 3% to $\pm 600 \text{ Oe}$ at 6%—attributable to the growing contribution of inter-cluster dipolar interactions and exchange coupling as Fe particle density increases, both requiring larger applied fields to overcome internal demagnetizing forces and achieve full moment alignment. While the 3% and 4% sample loops are nearly closed at the field origin (near-zero

remanence), the 6% sample shows a marginally wider loop opening, reflecting its slightly larger coercivity and remanence from stronger inter-particle exchange interactions at higher Fe loading. Figure 4 establishes the full magnetic fingerprint of this CVD-synthesized Fe-doped graphene series and provides the quantitative basis for the parameter extraction and structure–property analysis.

3.2 Saturation magnetization

Figure 5 shows saturation magnetization (M_s , $\text{emu} \cdot \text{g}^{-1}$) as a function of Fe doping concentration for CVD-grown graphene, with three data points at 3%, 4%, and 6% connected by straight lines. M_s increases monotonically with Fe content, consistent with the hysteresis loop data: from approximately $5 \text{ emu} \cdot \text{g}^{-1}$ at 3% to approximately $8 \text{ emu} \cdot \text{g}^{-1}$ at 4% and approximately $10 \text{ emu} \cdot \text{g}^{-1}$ at 6%—a twofold total increase across the doping range studied. The increase is non-linear: the slope between 3% and 4% Fe ($\alpha_1 \approx 3 \text{ emu} \cdot \text{g}^{-1}$ per 1% Fe) is substantially steeper than between 4% and 6% ($\alpha_2 \approx 1 \text{ emu} \cdot \text{g}^{-1}$ per 1% Fe). This is not simply a dilution effect. At lower Fe concentrations (3–4%), the majority of incorporated Fe species most likely occupy isolated adatom or substitutional positions within the graphene lattice, where each Fe atom contributes its full local magnetic moment ($\sim 3\text{--}4 \mu_B$ per atom) to M_s with minimal inter-site magnetic dilution. As the concentration rises from 4% to 6%, nucleation and growth of Fe nanoclusters at graphene grain boundaries and defect sites become increasingly prevalent. Within these clusters, antiferromagnetic exchange coupling between nearest-neighbour Fe atoms progressively cancels a fraction of the individual atomic moments, reducing the incremental gain in M_s per additional Fe atom. This crossover from single-ion-dominated to cluster-dominated magnetism with increasing Fe loading is a physically coherent explanation for the non-linear M_s –concentration relationship in Figure 5 (Zhang et al., 2011; Mathew et al., 2014).

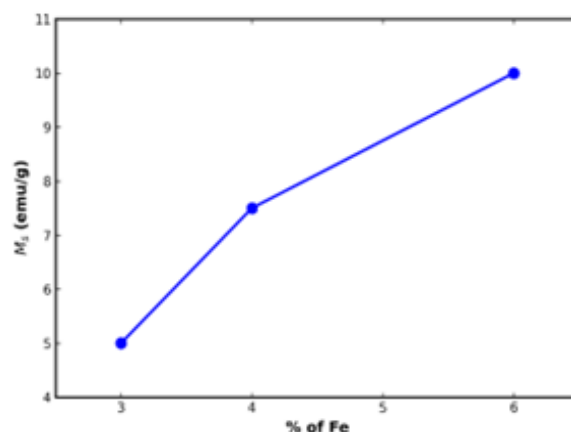


Figure 5. Saturation magnetization (M_s , $\text{emu} \cdot \text{g}^{-1}$) as a function of Fe doping concentration (3%, 4%, 6%) for CVD-grown Fe-doped graphene, extracted from room-temperature VSM hysteresis loops. The non-linear trend reflects a transition from single-ion to cluster-dominated magnetic interactions

3.3 Remanent magnetization

Figure 6 shows remanent magnetization (M_r , $\text{emu}\cdot\text{g}^{-1}$) of CVD-grown graphene as a function of Fe doping percentage, with data for 3%, 4%, and 6% doping levels connected by lines. M_r increases with Fe content: from approximately $0.1 \text{ emu}\cdot\text{g}^{-1}$ at 3% to approximately $0.3 \text{ emu}\cdot\text{g}^{-1}$ at 4% and approximately $0.4 \text{ emu}\cdot\text{g}^{-1}$ at 6%. While the absolute M_r values are modest in all three cases—consistent with the soft magnetic, low-anisotropy character established from the hysteresis loop analysis—the systematic increase with Fe loading is statistically robust and physically significant.

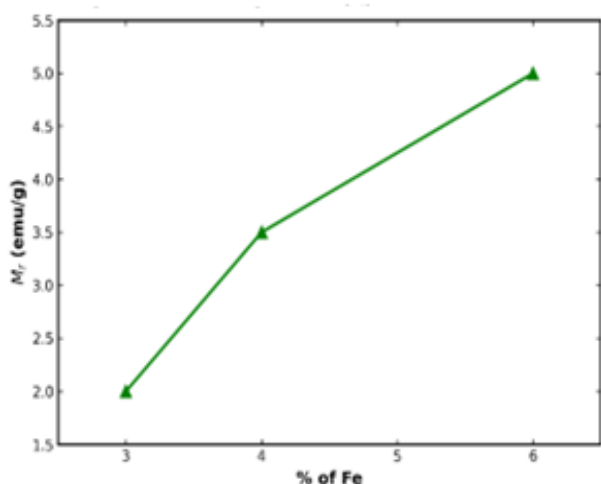


Figure 6. Remanent magnetization (M_r , $\text{emu}\cdot\text{g}^{-1}$) as a function of Fe doping concentration (3%, 4%, 6%) for CVD-grown Fe-doped graphene, extracted from room-temperature VSM hysteresis loops. The non-linear increase reflects the evolving magnetic coupling regime with increasing iron loading.

Like M_s , the M_r increase is non-linear, with a steeper slope between 3% and 4% than between 4% and 6%, again pointing to a shift in magnetic interaction regime with iron concentration. The squareness ratio (M_r/M_s), a normalized measure of magnetic memory independent of total moment, yields values of approximately 0.02, 0.04, and 0.04 for the 3%, 4%, and 6% samples, respectively—all well below the theoretical Stoner–Wohlfarth single-domain limit of 0.5, confirming the collective soft magnetic, multi-domain character of these materials. The slight increase in M_r/M_s between 3% and 4% parallels the onset of inter-cluster exchange coupling at higher Fe loading, which introduces additional magnetic pinning sites and stabilizes a slightly larger remnant domain configuration upon field removal. The near-constant squareness ratio between 4% and 6% is fully consistent with the reduced incremental increase in M_s in this concentration range, as discussed in Section 3.2. From a data storage perspective, while the present squareness ratios are insufficient for conventional permanent magnet

or hard disk write-head applications, the tunable M_r values and the clearly established concentration-dependent trend provide a route toward higher-remanence Fe-doped graphene composites through post-synthesis magnetic annealing, applied field cooling, or the introduction of magnetic anisotropy via substrate strain engineering or nanostructured pattern templates (Son et al., 2006; Yazyev, 2010).

4. CONCLUSION

Fe-doped graphene grown by CVD at 3%, 4%, and 6% iron content shows soft ferromagnetic ordering at room temperature across the full concentration range studied. The evidence is unambiguous: S-shaped M–H hysteresis loops with low coercivity ($H_c \approx 0.4\text{--}0.8 \text{ Oe}$) and low remanent magnetization ($M_r \approx 0.1\text{--}0.4 \text{ emu}\cdot\text{g}^{-1}$) appeared consistently across all three compositions, and saturation magnetization climbed from $M_s \approx 5 \text{ emu}\cdot\text{g}^{-1}$ at 3% Fe to $M_s \approx 10 \text{ emu}\cdot\text{g}^{-1}$ at 6% Fe a clean twofold increase that makes iron concentration an effective and predictable handle on magnetic output.

The concentration property relationship is not simply linear, and that non-linearity carries physical meaning. The steeper M_s and M_r gains between 3% and 4% Fe, compared to the more modest gains between 4% and 6%, reflect a transition in the dominant magnetic interaction regime: isolated Fe adatoms and substitutional dopants at low loading give way progressively to Fe nanoclusters at grain boundaries and defect sites as concentration rises, and intra-cluster antiferromagnetic exchange coupling in those clusters partially cancels the individual atomic moments. The crossover is visible in the data and consistent with DFT predictions and prior experimental work on transition metal-doped graphene systems. It also sets a practical ceiling: beyond a certain Fe loading, adding more iron returns diminishing magnetic gains.

What these results establish, taken together, is a quantitative and mechanistically grounded map of how magnetic performance scales with doping in CVD-grown Fe-doped graphene. The 6% Fe sample delivers the strongest ferromagnetic response and is the natural starting point for high-sensitivity sensor and spintronic transducer design. The soft magnetic character shared across the series low squareness ratio, easy reversal, superparamagnetic contributions at lower doping suits the material well to STT-MRAM free layers and RF spintronic inductors. Where higher remanence or coercivity is required, the path forward is clear: field annealing, substrate strain engineering, or co-doping with Co or Ni to introduce controlled magnetic anisotropy. The CVD synthesis protocol and the concentration-dependent magnetic dataset reported here provide the quantitative foundation on which that further engineering can proceed.

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