

Structural and magnetic properties of Fe-membranes embedded in hexagonal graphene nanoholes

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ABSTRACT

The structure and magnetism of Fe-doped hexagonal graphene nanoholes (h-GNHs) were studied on the basis of density-functional theory. The nonzero magnetic moment of carbon in zigzag edge can be greatly reduced in Fe-doped h-GNHs due to the local C–Fe interaction. The reduced magnetic moment is almost zero, and its direction is opposite to that of iron atoms. Therefore, Fe-adsorption may be used to destroy the spin polarization of graphene zigzag edge. In Fe-doped h-GNHs, iron atom would prefer to adsorb on graphene armchair edge and form five-member (1Fe + 4C) ring. In stable Fe-doped h-GNHs, the bond length of C–Fe is about 1.9 Å. And in the most stable Fe-membrane embedded h-GNH with the largest binding energy, the average Fe–Fe bond length is about 2.36 Å. Besides, the mean magnetic moment induced by iron atoms changes greatly from 1.45 μ_B per iron atom to 3.46 μ_B per iron atom in Fe-doped h-GNHs. The magnetism of iron seems to be sophisticated. It is related to the type of graphene edge, the size of GNH and the number of iron atoms.

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1. Introduction

Magnetism of carbon materials is of particular interest since magnetism is commonly associated with elements containing *d* or *f* electrons [1–4]. Ideal and intact graphene is nonmagnetic. However, graphene nanoribbons, nanoflakes and nanoholes with zigzag edges have been shown to exhibit magnetism, which is from the localized edge states [5–11]. Magnetism of graphene induced by defects was also studied in many works [12–14]. Moreover, it has been reported that structural distortions can be observed when multi-atom vacancies are created in graphene [15,16]. Interestingly, the presence of doping can reduce the probability of distortion and coalescence of vacancies in graphene [17–20].

The magnetism of graphene can also be induced by magnetic proximity effect [21,22]. Especially, there is a strong graphene–Fe interaction when iron atoms are intercalated in the interface of graphene/substrate [23,24]. The intercalation of iron atoms between graphene and Ni(1 1 1) can change drastically the magnetic response from the graphene layer [25]. Similarly, the intercalation of iron atoms through graphene formed on SiC(0 0 1) surface was found to induce significant changes in the electronic properties of graphene due mainly to the Fe-induced asymmetries

in charge as well as spin distribution [21]. Moreover, graphene–substrate interaction can be fine-tuned by Fe–Ir alloying at interface [22]. When a critical Ir-concentration close to 0.25 is reached in the Fe layer, a transition in graphene layer from chemisorption to physisorption takes place. Furthermore, the transition is accompanied by an inversion of the induced magnetization of graphene due to the coupling with the iron atoms from antiferromagnetic coupling to weakly ferromagnetic coupling.

Besides, two-dimensional (2D) materials have also received much attention in experiment and in theory since the successful invention of graphene [26–29]. Especially, free-standing single-atom-thick iron membranes were realized in experiment [30], although the excess of surface dangling bonds makes the formation of free-standing 2D metals unstable and hence difficult to achieve. The 2D metal magnets may be promising for magnetic recording media [30]. In experiment, iron atoms can be found as small nanocrystal forming on the surface of graphene, as single atom or small cluster at the edge of pores in clean graphene, or as 2D crystalline membrane suspended across perforations in graphene. Although the embedded iron membrane in graphene perforations has square lattice configuration in experiment [30], free-standing monolayer iron may be more stable in triangular lattice compared to square lattice [31,32]. As for the magnetism of the 2D metal, the magnetic moment of iron atom is markedly higher than that in its bulk counterpart. Theoretically, strain may lead to dramatic

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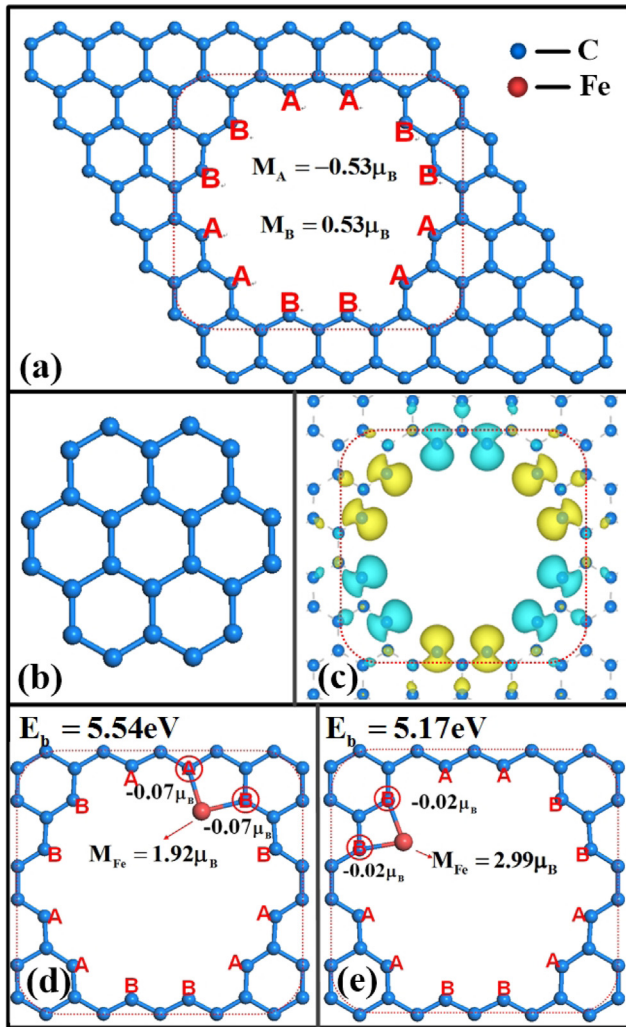


Fig. 1. (a) The structure and magnetism of h-GNH with symmetric 24-order vacancy (24-GNH). (b) The complementary part of the removed 24 carbon atoms. (c) The spin charge density of the 24-GNH. The yellow indicates spin up and the cyan indicates spin down. (d, e) The structure and magnetism of single-Fe-doped 24-GNH. The iron atom is at AE in (d) and at ZE in (e). The binding energy is about 5.54 eV (d) and 5.17 eV (e), respectively. The carbon atoms at edge are labeled by A and B to identify the different sublattices in graphene. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

changes in the magnetic configurations for both small iron cluster and single layer iron membrane embedded graphene perforations [33,34]. It may be an easy and effective method to introduce and tune the magnetic properties of graphene, and offer a new direction for the development of graphene-based spintronic devices.

In this work, we use the first-principles calculation based on density-functional theory (DFT) to study the structure and magnetism of Fe-doped hexagonal graphene nanoholes (h-GNHs). The considered h-GNHs have D_{6h} symmetry and do not exhibit

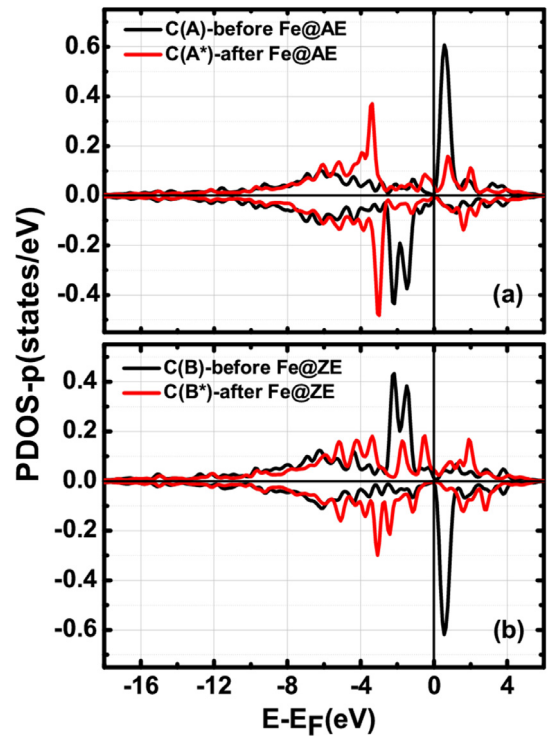


Fig. 2. Partial density of states of carbon p-orbital (PDOS-p) before and after Fe-adsorption in 24-GNH. The single iron atom is at AE (a) and ZE (b), respectively. The carbon atoms are the nearest carbon atoms from iron atom at edge, which are corresponding to the carbon atoms selected in Table 1.

evident distortion and coalescence [16]. In these structures, the armchair edge and the zigzag edge can be studied symmetrically. And the possible structure of Fe-doped h-GNHs would be simplified in a degree. Besides, considering the symmetry of h-GNHs, the doped iron atoms are mainly located in triangular lattice. Triangular lattice is also more stable for free-standing monolayer iron [31,32].

2. Computational details

The total energy, stable structure and magnetism were calculated on the basis of DFT using the projector augmented wave (PAW) method implemented in the Vienna Ab-initio Simulation Package (VASP) [35]. In all calculations, it was used that the generalized gradient approximation (GGA) method for the exchange-correlation [36]. A 7×7 graphene supercell was considered for different sizes of GNHs. Such supercell is large enough to avoid the interactions between GNHs. The lattice constant of the supercell is set to be $7 \times 2.46 \text{ \AA} = 17.22 \text{ \AA}$. The structure was modeled by periodic slab geometry, with a vacuum of at least 10 \AA between two neighboring slabs. The total energy was converged to better

Table 1

The magnetic moment, the total charge and the transferred charge of carbon atom at edge before and after single iron atom doping in 24-GNH. There are two nearest carbon atoms from iron atom in each adsorption configuration. Symmetrically, we choose one of them to consider. The carbon atoms are marked in circle in Fig. 1(d) and (e).

Title	Before and after(°) Fe@AE		Before and after(°) Fe@ZE	
	C(A)	C(A*)	C(B)	C(B*)
Magnetic moment (μ_B)	-0.53	-0.07	0.53	-0.02
Total charge (e)	2.53	2.56	2.52	2.56
Transferred charge (e)	0.03		0.04	

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