Tunable electronic structure and spin splitting in single and multiple Fe-adsorbed g-C_2N with different layers: A first-principles study

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Abstract

The electronic structure of Fe adsorbed g-C_2N with different layers is investigated by first-principles calculations. The Fe\(^{1}\) and Fe\(^{2}\) represent the Fe adsorptions at C=C and C=N rings, and Fe\(^{11}\) and Fe\(^{121}\) adsorption sites are also considered. The Fe\(^{1}\) adsorbed g-C_2N is metallic with layer from n=1 to 4, and the maximum spin splitting is 515, 428, 46 and 133 meV. The band gap of Fe\(^{2}\) adsorbed g-C_2N with different layers is 0, 0, 117 and 6 meV, and the maximum spin splitting is 565, 369, 195 and 146 meV, respectively. All of the Fe\(^{11}\) adsorbed g-C_2N are metallic with layer from n=1 to 4, and the maximum spin splitting is 199, 83 and 203 meV. An indirect band gap of 215 meV appears in Fe\(^{121}\) adsorbed g-C_2N at layer n=3, and the maximum spin splitting is 283, 211, 304 and 153 meV, respectively. Our results show that the electronic structures of Fe adsorbed novel two-dimensional semiconductor g-C_2N can be tuned by different layers. Moreover, the spin splitting of Fe\(^{2}\) adsorbed g-C_2N decreases monotonically as g-C_2N layer increases from n=1 to 4, which will provide more potential applications in spintronic devices.

1. Introduction

Since the discovery of graphene, owing to its unique two-dimensional (2D) layered structure and the extraordinary electronic properties with a Dirac cone, a wide range of research enthusiasm on 2D materials have been provoked [1–3]. Recently, 2D materials have exhibited better properties than conventional materials, which are potential candidates for the next generation electronic, optoelectronic and spintronic devices [4–6]. However, graphene also has its limitations, such as a small band gap, which seriously restricts its practical application [7]. Therefore, finding a new 2D material with a large band gap is necessary [8]. A new graphene-like semiconductor g-C_2N is successfully synthesized by wet chemical method [9]. The monolayer g-C_2N is a semiconductor with a direct band gap of 1.66 eV and is maintained as a direct band gap semiconductor at strain, the band gap can be tuned as large as 1.00 eV [10,11]. When the g-C_2N serves as a substrate, graphene can open a band gap of 0.24 eV and the g-C_2N/graphene heterostructure has a large on/off ratio, which makes it possible to produce a specific application in field effect transistors [10,11]. A similar 2D semiconductor g-C_2N\(_4\) has a 1.10 eV indirect band gap, and its internal atoms are only distributed in half of lattice [12]. Thus, g-C_2N has a greater band gap and better symmetry, presenting more potential applications in electronic and optoelectronic devices [9–11].

Additionally, the transition metal (TM) atoms are considered to be the source of magnetism. It is found that the TM adsorbed on h-BN\(_2\) is a magnetic semiconductor at Fe adsorption, which is very interesting [13]. In TM adsorbed germanene, most of the adsorbed structures turn into metallic properties, but in case of Ni and Zn adsorption are semiconductors, where the largest spin splitting and magnetic moment appear at Fe adsorption [14]. A similar phenomenon also appears in TM adsorbed 2D silicon carbide [15]. The TM adsorption can result in total magnetic semiconductor at Fe adsorption, which is very interesting [13].

In spintronics, the aim is to utilize the degree of freedom of spin in logic and memory devices [18,19]. The TM adsorbed structure is beneficial to apply spin splitting to magnetic electronic devices [20,21]. The electronic structure of Fe-adsorbed graphene has been predicted to be effective for hydrogen storage [22,23], and Fe-adsorbed graphene can...
effectively catalyze formaldehyde decomposition and CO oxidation at room temperature [24,25]. Therefore, studying the electronic structure of Fe-adsorbed g-C2N has significant implications for the design of catalysts and electronic devices. In this work, the electronic structure of Fe adsorbed g-C2N with different layers is studied by first-principles calculations, where the effects of different adsorption sites of Fe atoms are also considered. The spin splitting and magnetic moments in Fe adsorbed 2D semiconductor g-C2N with different layers appear. Moreover, the spin splitting of Fe adsorbed g-C2N at C–N ring decreases monotonically as g-C2N layer increases from \( n = 1 \) to 4, which will provide more potential applications in spintronics devices.

2. Computational methods

The electronic structure of single and multiple Fe adsorbed g-C2N with different layers are performed by Vienna ab-initio simulation package, which is based on the density functional theory (DFT) with Coulomb correction [26–28]. Conventional GGA does not work well with experimental results when the magnetic structure is considered because TM 3d orbitals are sensitive to Coulomb potential, so it is imperative to use the GGA + \( U \) method to correct these defects [29,30]. The initial magnetic moment of Fe is 4 \( \mu_B \) [31]. By considering the finite onsite Coulomb interaction of Fe d electrons, the method of DFT + \( U \) makes a key simplification

\[
U_{\text{eff}} = U - J
\]  

where the \( J \) is usually assumed to be 0.00 eV to simplify expression. So, the “+\( U \)” is used for Coulomb correction [14,29], where the value of \( U \) on TM adsorption structures is selected as 4.00 eV [30]. The calculation of exchange-correlation potential is realized by generalized gradient approximation with Perdew-Burke-Ernzerhof (PBE) functional [32,33]. A \( \Gamma \)-centered 5 \( \times \) 5 \( \times \) 1 Monkhorst-Pack k-point is set in Brillouin zone, and the plane wave cut-off energy is set at 500 eV [34]. In order to avoid the influence of periodic structure, a 16-Å vacuum layer is added in the \( z \) direction [35]. All of the Fe adsorbed g-C2N with different layers are relaxed by DFT + \( U \) until the forces are converged to \( 10^{-2} \) eV/Å and the energy to \( 10^{-5} \) eV [36].

The lattice constant of g-C2N is 8.330 Å and the length of C–N bond is 1.337 Å, which is consistent with previous reports [9,11]. After optimization calculations, Fe adsorbed g-C2N with different layers only can be converged at \( n = 1, 2, 3 \) and 4 layers. The Fe\(^1\) and Fe\(^2\) are defined as Fe atoms adsorbed on the C–C and C–N rings, respectively. So the Fe\(^{11}\) represents multiple Fe adsorbed at C–C rings. In order to reveal the actual g-C2N layer effects on adsorption structures, it can also be studied from the gain and loss of electrons [37,38]. The Fe atoms and g-C2N layers are cleaved from the adsorption structure separately to calculate the charge density difference

\[
\Delta \rho = \rho_{\text{Fe-C2N/TM}} - \rho_{\text{Fe-C2N}} - \rho_{\text{TM}} 
\]  

Fig. 1. The top and side view of single Fe adsorbed g-C2N with \( n (n = 1, 2, 3 \) and 4) layers, (a) Fe\(^1\) and (b) Fe\(^2\) adsorption sites. Fe\(^1\) and Fe\(^2\) represent Fe adsorbed at C–C and C–N rings, respectively.

Fig. 2. The top and side view of multiple Fe adsorbed g-C2N with \( n (n = 1, 2, 3 \) and 4) layers, (a) Fe\(^{11}\) and (b) Fe\(^{121}\) adsorption sites. Fe\(^{11}\) represents multiple Fe adsorbed at C–C rings.
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