First Principles Study of spin polarization in Fe−doped monolayer C2N−*h*2D

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Recent multifunctional two-dimensional material research has triggered huge interests in various modifications for substitution of atoms. Instead of novel metals used as the most popular catalysts, nonprecious transition metals are promising candidates for efficient oxidation-reduction transfers. The recent discovery of Co@C_2N has an alternate possiblity as catalysts for the ORR(Oxygen Reduction Reaction) in DSSc(Dye Sensitized Solar Cell) and OER(Oxygen evolution cobalt oxides). Here we report spin-polarized DFT calculations of the structure doped Iron that is one of ferromagnetism atoms like Co to provide a basic desciption of the ferromagnetism of the elemental metals. The spin-density-funtional results present the most stable state energetically is when having pairwise up/down spin.

INTRODUCTION

The recent research on the design of a two-dimensional crystal with uniform holes and nitrogen atoms as multifunctional two-dimensional material has garnered interests in many areas. The nitrogenated holey two-dimensional crystal structure has potentional for multifunctional 2D crystals as practical applications.[1] What is more, cobalt oxide nanoparticles encapsulated in C_2N-*h*2D network polymer (Co@C_2N) has experimently evaluated as a promising candidate for a novel catalyst as to the hydrogen evolution and hetergeneous reduction.[2] Instead of

novel metals, inexpensive materials can have great potential to be used as catalysts with ferromagnetism atoms like Co, Fe, and Ni.

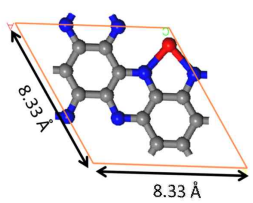
Here, we carried out spin-polarized density-functional theory(DFT) calculations to understand the magnetism of itinerant electrons in Fe-doped monolayer C_2N-*h*2D.

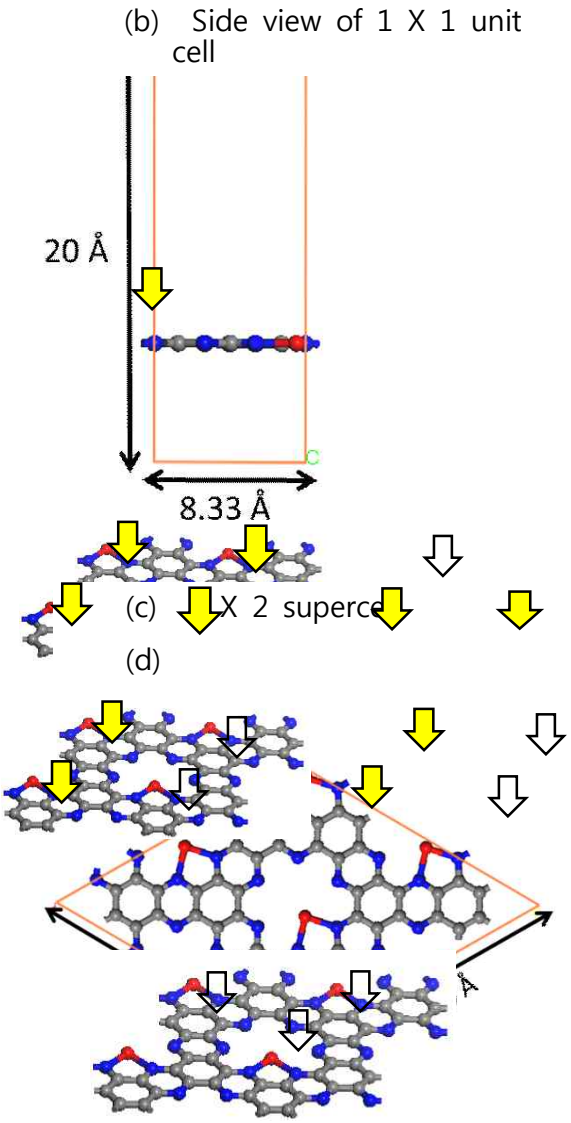
COMPUTATIONAL METHODS

In this study, all the geometry optimization and calculation we carried out are based on the spin-polarized density functional theory(DFT) using the EDISON DFT software in LCAODFTLab of the EDISON Nano-physics. The local density approximations and Caperley-Alder(LDA-CA) are used. In all calculations, these were performed with 200 Ry cutoffs. For the geometry optimization, 5 × 5 × 1 Monkhorst−Pack grid *k*-meshe is adopted for a single-layer C_2N-*h*2D doping Iron. These structures are fully relaxed until converged to 1.0^{-4} eV and 0.02 eV/Å, energy and forces respectively.

Fig. 1. Theoretical modeling of the atomic structure of monolayer Fe@C_2N : (a)Top view, (b)side view and (c) 2 X 2 supercell.

Red ball and blue ball mean Fe and Ni,respectively. And the other is C.





RESULTS AND DISCUSSION

First of all, we explored the geometric properties and electronic structure of monolayer Fe@C_2N. The atomistic ball-stick models of monolayer Fe@C_2N are optimized as illustrated in Fig. 1. The lattice parameters of monolayer Fe@C_2N is equivalently 8.33 Å, generating the covalent bond lengths as C-C of 1.385/1.485 Å, C-N of 1.345/1.381 Å, and N-Fe of 2.003 Å.

We have performed the total energy calculations to find the most stable state among spin-polarization differences. In the case of 1 X 1 unit cell, the initial spin polarization of Fe is 4 u B /atom. while total B /atom H1 eV. all of the possible case to change spin directions in 2 X 2 supercell. As illustrated in **Fig 2** , there are 4 cases for a pair of coupled spins. Moreover there is only one case for unpairwise spin direction like up, down, down, and down.

(a) Top view of 1 X 1 unit cell

Fig. 2. The number of occasion depended on spin directions

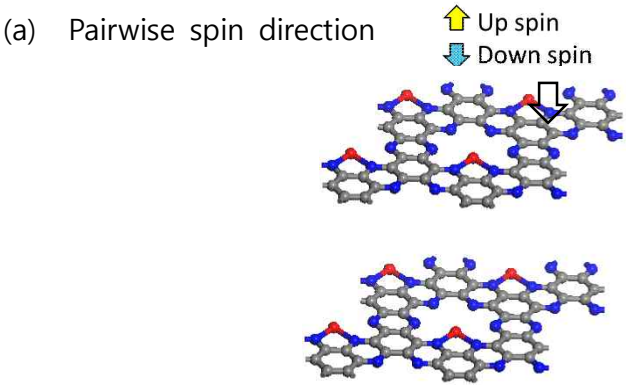


Table 1. Total energies of Spin plarized calcultions : (+) means up and (–) means down.

(a) Pairwise spin direction

Spin direction	Initial spin polarization	Total spin polarization	E_{tot} (eV)
++++	16	15.181124	-16088.329400
++--	0	0	-16088.329638
+-+-	0	0	-16088.325815
+--+	0	0	-16088.325694

(b) Unpairwise spin diection

Spin direction	Initial spin polarization	Total spin polarization	E_{tot} (eV)
+---	-8	-7.650727	-16088.249402

Resultingly, the more stable state is when spin directions are pairwise than unpairwise as shown in **Table 2**. However, it is hard to

predict what state the most stable is in a pair of coupled directions. This is because these energies differences have possibility by some conditions like temperature etc. Comparatively, the stable state would be when all the spin is up or the second condition of **Fig. 2(a)** than others.

CONCLUSION

We investigated Total energies of Fe-containing C_2N-*h*2D structure spin-polarized DFT calculations to find out the most stable state with differences of spin directions.

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