DFT SIMULATIONS OF HYDROGEN ADSORPED ON SILICON CARBIDE NANOSHEET

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Abstract

The Density Functional study on Hydrogen Adsorption on Silicon Carbide Nanosheet has been investigated. All calculations have been performed with density functional theory (DFT) , within the plane-wave pseudopotential approach as implemented in the Quantum ESPRESSO Simulation Package. The Perdew-Burke-Erenzerhof (PBE) formulation of the generalized gradient approximation (GGA) was employed to describe the exchange and correlation energies. we've tried different positions for hydrogen adsorption after simulating our honeycomb nanosheet of SiC. Four different positions of adsorptions are considered in the survey and it is finally shown that the most stable state happens when hydrogen atoms are adsorbed on silicon and carbon atoms at the two opposite sides of silicon carbide nanosheet. This adsorption have made some changes in the atoms positions so that the nano sheet didn't remain flat any more. The results have shown that this structure was the most stable one among those four. Silicon carbide is a semiconductor with a wide band gap about 2.5851 eV. After the hydrogen adsorption, the hydrogenated silicon carbide nano sheet have has a band gap about 3.9499 eV, which is much more than the band gap in the pure structure.

Keywords: DFT, silicon carbide, nano sheet, hydrogen adsorption

1. INTRODUCTION

Hydrogen has been identified as one of the future energy carrier due to several inherent advantages including high energy density, light weight, no CO2 emission, and abundant nature [1-5]. For the last three decades the efforts has been taken to find out suitable hydrogen storage materials with high hydrogen storage capacity and good hydrogen adsorption behavior. Among the various solid state options available, carbon nanostructures have been studied extensively due to its light weight, diversity in structures, large surface area and interesting hydrogen adsorption properties. Apart from the carbon nanostructure, another very promising hydrogen storage alternative is Si/SiC nanostructure [6-8]. Si being in the same group with the carbon it shows properties quite similar to that of C. Again, as the polarizability of Si is more than C, so it is expected that, due to stronger van der Waal's interaction, SiC/Si nanostructures can bind hydrogen more strongly compared to the pure carbon nanostructures. Mpourmpakis et al, 2006 [9] have proposed that pure SiC nanotube can improve the binding energy with hydrogen molecule by 20% compared to pure carbon nanotube.

Unlike the polymorphs of carbon, SiC is a polar material. In spite of the fact that both constituents of SiC are Group IV elements, charge is transferred from Si to C due to higher electronegativity of C relative to Si atom. Silicon carbide is a binary compound of carbon and silicon as widely used in electronic devices.

2. RESULTS AND DISCUSSION

The density functional theory (DFT) [10, 11] calculations were carried out using the Quantum Espresso Package. The Perdew-Burke-Erenzerhof (PBE) [12, 13] formulation of the generalized gradient approximation (GGA) was employed to describe the exchange and correlation energies. For the SiC graphene-like sheet calculation the sheet has been modeled by alternatively arranged 16 C and 16 Si atoms. We employed the two dimensional periodic boundary condition in the layered plane with a vacuum region (15A°) between sheets to ensure there was no interaction between SiC sheets. In order to establish the accuracy of the present

method we have performed few test calculations of SiC nano sheet using several sets of k-points according to the Monkhorst-Pack scheme [14] so as to ensure the convergence of the total energy. Finally the k-points were taken as 7×7×1. We've also found the optimized values of e-cut as 80 Ry. This optimization was made for the unit cell of the SiC nano sheet (consists of two atoms; one Si and one C) which can be generalized to the main structure. In **Figure 1** we have shown the optimized structure of the SiC nano sheet from top view and side view.

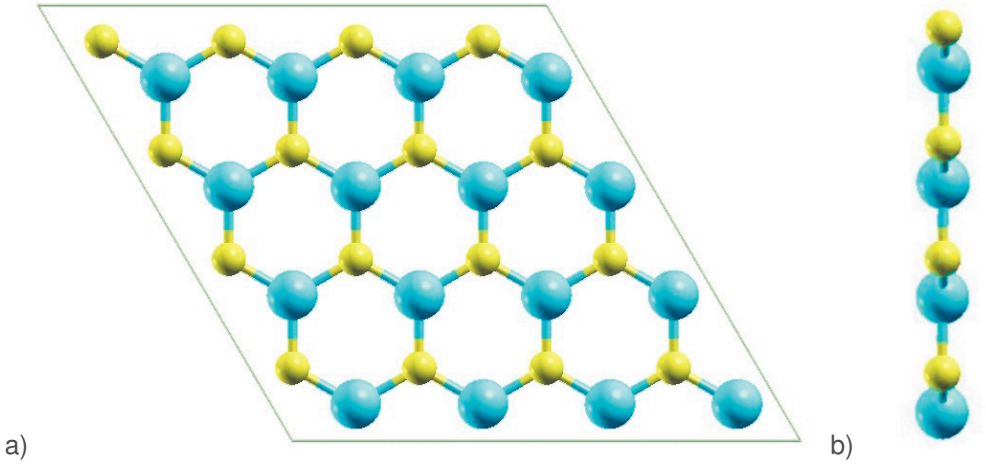

Figure 1 (a) Top view and (b) side view model of pure SiC sheet. Blue and yellow balls represent Si and C atoms, respectively

In the optimized geometry, all the Si-C bonds are found to be identical. The minimum of total energy occurred when Si and C atoms are placed in the same plane forming a honeycomb structure. The calculated Si-C bond length of the sheet is about 1.7788 Å and the Si-C-Si angle is 120° which are in a good agreement with previous calculations and experimental reports [15,16]. The magnitude of the Bravais lattice vectors of the hexagonal lattice is found to be $a_1=a_2=3.080$ Å.

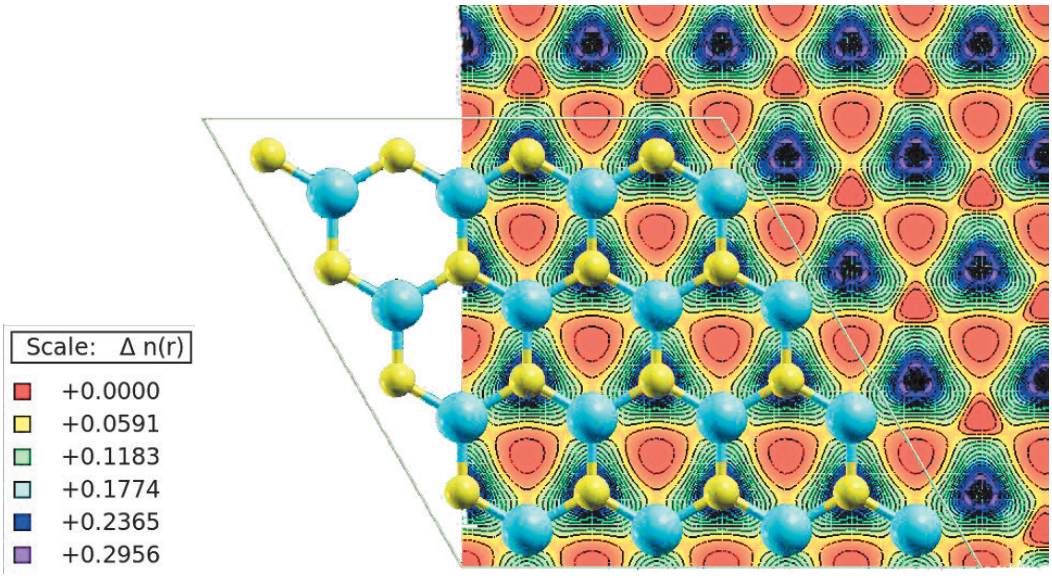

Figure 2 The charge density distribution on the SiC nano sheet. Figure shows higher charge on the C atom and lower charge on the SiC atom

From the charge density distribution of the SiC sheet (**Figure 2**), it is seen that a considerable amount of electronic charge is transferred from Si to C site. The presence of these point charges on the SiC sheet influence the hydrogen adsorption properties.

In order to study the electronic properties of the structure, the results of the band structure diagram is important, because we can derive the conductive properties of the structure out of it. The Band Structure of the silicon carbide nanosheet is shown in **Figure 3**. The results show that the considered structures band gap is about 2.5821 eV which indicates that it is a semiconductor with approximately large band gap. This result is very close to the reported band gap [17].

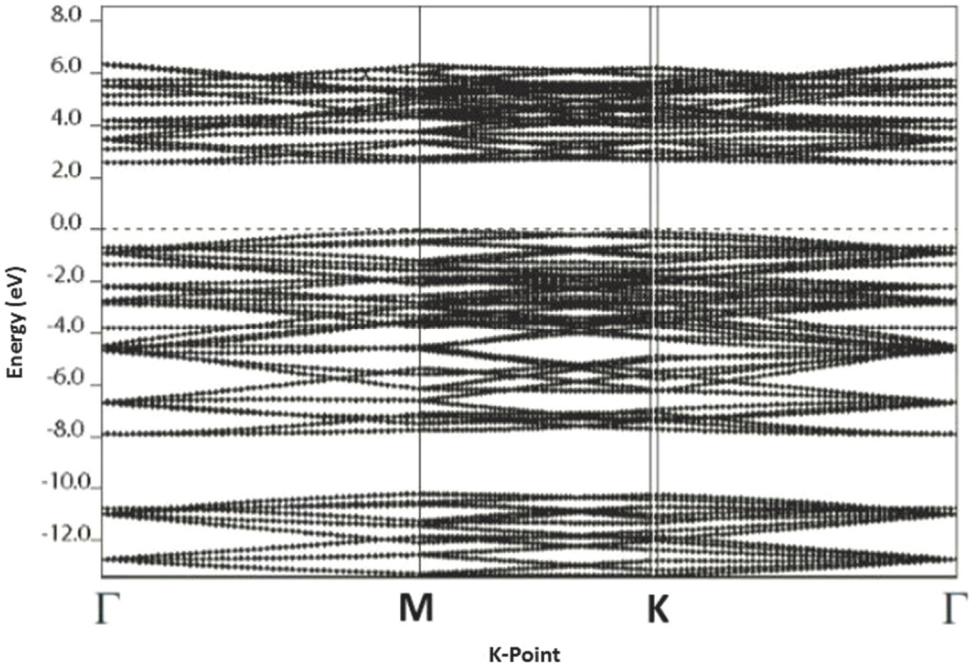

Figure 3 Band structure of the pure silicon carbide nano sheet

After establishing the pure structure of SiC nano sheet, we attempt to investigate electronic properties of the nano sheet under hydrogen adsorption. Four different positions for hydrogen adsorption are considered: (1) adsorption of hydrogen atoms on silicon atoms, (2) adsorption of hydrogen atoms on carbon atoms, (3) adsorption of hydrogen atoms on silicon and carbon atoms at the two opposite sides of SiC sheet (4) adsorption of hydrogen atoms on silicon and carbon atoms at the same side of SiC sheet. It is vital to say that the first and second states were not convergented so we assumed they were not stable states. As a result, we continued our work on the last two states. State (3) have had a total energy about -346.12 Ry, on the other hand, state (4) have had a total energy about -344.16 Ry. Therefore we picked state (3) as the most stable state for hydrogen adsorption; adsorption of hydrogen atoms on silicon and carbon atoms at the two opposite sides of SiC sheet. The Figures of these two states are shown below. It should be remarked that the relaxed structures are exactly shown in **Figure 4**.

After establishing the hydrogenated structure, we have studied the electronic properties of the silicon carbide nano sheet under hydrogen adsorption. For this purpose, we again refer to the band structure diagram. This diagram is so important, because we can derive the conductive properties of the structure out of it. The Band Structure of the silicon carbide nanosheet under hydrogen adsorption is shown in **Figure 5**. The results show that the considered structure's band gap is about 3.9499 eV which is much more than the pure silicon carbide nano sheet.

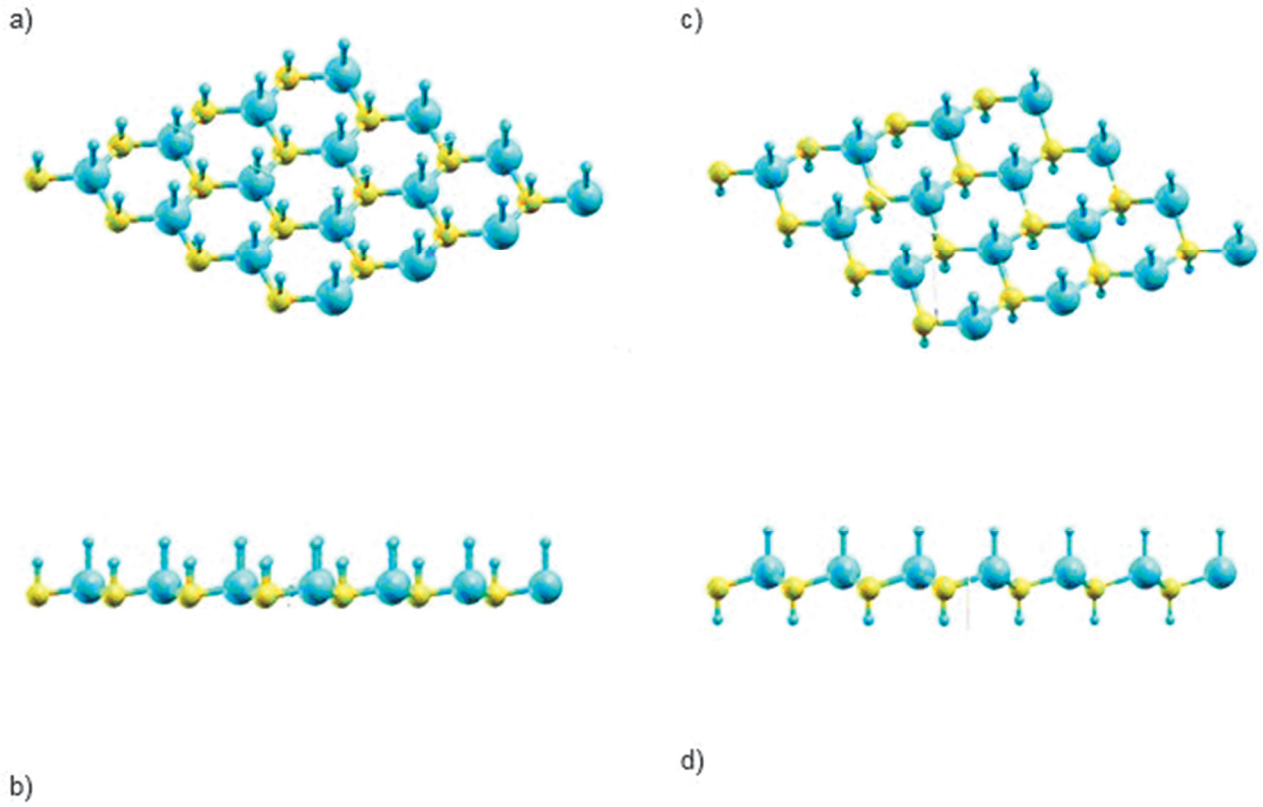

Figure 4 (a) Top view and (b) side view model of hydrogenated SiC nano sheet in state (4). (c) Top view and (d) side view model of hydrogenated SiC nano sheet in state (3) (Big blue and yellow balls represent Si and C atoms, respectively; little blue balls represent hydrogen atoms)

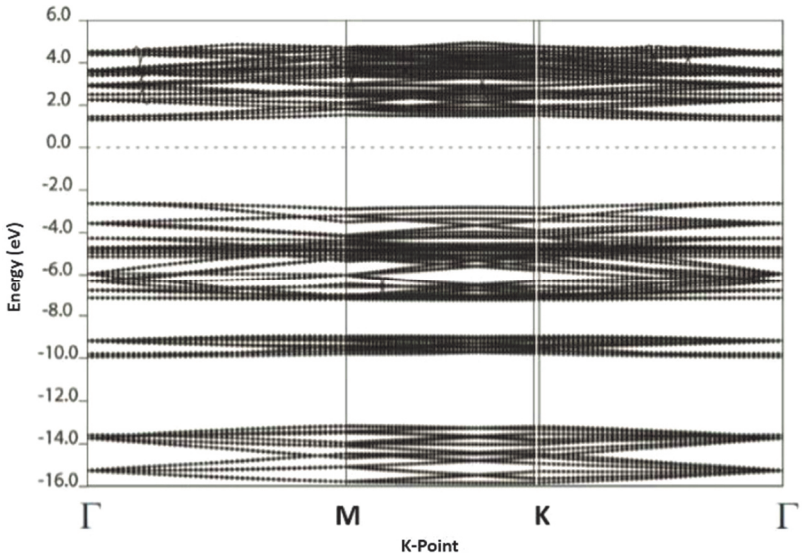

Figure 5 Band structure of the hydrogenated silicon carbide nano sheet

3. CONCLUSION

In this survey we've studied electronic properties of silicon carbide nanosheet under hydrogen adsorption based on the Density Functional Theory (DFT) and using Quantum Espresso simulation software. The results of these calculations have shown that the SiC nanosheet is a semiconductor with a band gap about 2.5821 eV.

This material has high strength and is widely used in electronic devices. We've also tried different positions on the SiC nano sheet for hydrogen adsorption. After testing several positions, it was concluded that the most stable state happens when hydrogen atoms are adsorbed on silicon and carbon atoms at the two opposite sides of silicon carbide hexagonal nano sheet. Finally, we calculated the band gap of the hydrogenated SiC nanosheet, which is about 3.9499 eV.

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