Density functional theory simulation of nitrogen-vacancy center in diamond as quantum sensor for neutrino mass detection

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ABSTRACT

Nitrogen-vacancy (NV) pair in diamond has been widely investigated due to its profound quantum properties even at relatively higher temperature compared with those materials having to satisfy strict cryogenic conditions to display quantum states. Spin properties of NV center in diamond can be tuned by external electromagnetic radiations, which leads to its application in detecting microwave signals. Here in this work, density functional theory (DFT) method has been used to analyze electronic properties of diamond with NV centers with the aim of utilizing it for detecting very weak microwave radiations from electron cryotron radiations, to ultimately determine neutrino mass.

Keywords: NV center in diamond, DFT, Neutrino mass detection

1. INTRODUCTION

The nitrogen-vacancy (NV) center in diamond has attracted much research interests due to its capability of demonstrating quantum states at room-temperature [1]. By substitutionally doping a nitrogen atom and removing an adjacent carbon atom a nitrogen-vacancy pair is formed. The negatively charged NV pair has single-photon absorption (typically green light) ability and subsequently emits visible light (typically near infrared). The quantum properties of the NV center can be affected by various external parameters, such as electric field, microwave radiation, light, etc. This leads to its potential application in neutrino mass determination in the CRESDA (cyclotron radiation electron spectroscopy demonstrator apparatus). The working mechanism is that the cyclotron electron emits microwave signal, which will be picked up from the PL (photoluminescence) measurement of the NV center in diamond. This _all-optical_ detection mechanism has benefits of low electronic noise, and high signal-to-noise ratio. In this work, density functional theory is used to analyze the band structures, optical absorption properties, and local density of states of the NV center. It is known that traditional generalized gradient approximation (GGA) and the parametrization of Perdew-Burke–Ernzerhof (PBE) lead to underestimation of the bandgaps. Hence in this work, the Hybrid-GGA with HSE06 is used to mitigate this issue [2]. Using the NV center in diamond for quantum sensing, signal detection depends on the dipole coupling with external signals, which decays with distance. Therefore, the NV center needs to be close to surface [3]. This paper starts building bulk cubic supercells of the pure diamond and defected by NV pairs, followed by DFT calculations on their band structures and optical absorption coefficients. Then a thin film model of the NV diamond has been built with fluorene passivation on both surfaces. Electronic properties such as band structures, density of states of the thin film model have been calculate and presented.

1. SIMULATION METHOD

The schematic graph of using NV diamond in the detection of neutrino mass is shown in Figure 1. It is seen that electron propagates along the magnetic field direction, emitting electromagnetic waves, which can be subsequently captured by the NV center in diamond. It is then detected by the PL measurement. Because there will be no electrical current being involved in the detection mechanism, this all-optical or 'cold' sensing method benefits from the ultra-low electronic noises, yielding much higher signal-to-noise ratios. To implement this method in the neutrino mass measurement, deeper understanding of the NV diamond assisted by the first principles method is essential.

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Starting with a pristine bulk diamond DFT supercell model that contains 64 C atoms with the crystal lattice type of simple cubic. The optimized lattice constant is 7.132 Å, and the space group is Fd-3m. Then the NV center was created by replacing a C atom with a N atom and removing a C atom adjacent to the N atom, which was geometrically optimized. The optimized NV diamond cell has 62 C atoms and 1 N atom, with the same lattice type. The lattice constant is 7.144 Å, and the space group is R3m. The geometrical optimization calculation was conducted with the force field calculator embedded in the QuantumATK software.
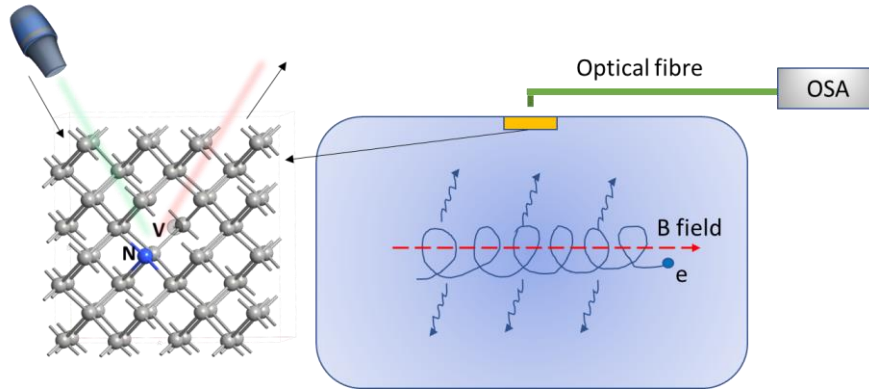

Figure 1. Schematic of using NV in diamond in CRESDA.

Calculations were conducted for both defected and pristine diamond models using the linear combination of atomic orbitals (LCAO) algorithm that builds up the Green function in the molecule. The exchange correlation potential was the Hybrid functionals that incorporate a portion of exact exchange from Hartree-Fock theory. The basic parameters used in the simulation for the electronic properties are as follows: broadening is 25 meV; density mesh cut-off is 60 Hartree; k-points sampling is set to 4x4x4. These parameters were used for calculating the electronic properties of both the pristine and defected diamond supercells. In the geometrical optimization, the maximum force tolerance was 0.01 eV/Å and maximum stress tolerance was 0.1 GPa, which were used for both pristine and defected models. Band structures and optical absorption coefficients have been calculated for both models described above (shown in Figures 2 & 3). In the thin film model, 20 layers of C atoms with the <110> cut surface were created, passivated by the fluorene atoms on both surfaces (shown in Figure 4). The supercell contains 358 C atoms, 36 F atoms and one N atom. The geometrical optimization was done using the generalized gradient approximation (GGA) with the Perdew-Burke-Ernzerhof (PBE) functionals. The k-point sampling was set to 4x3x1, density mesh cut-off was 80 Hartree, and the broadening was 200 meV. With regards to the optimization parameters, the maximum force tolerance was 0.01 eV/Å and the maximum stress tolerance was 0.1 GPa. In the property calculations, the Hybrid-GGA with HSE06 was used, which can have much more accuracy than the traditional GGA method. The parameters in the property calculations are as follows: broadening was 25 meV, density mesh cut-off was 80 Hartree, and the predefined functionals was HSE06.
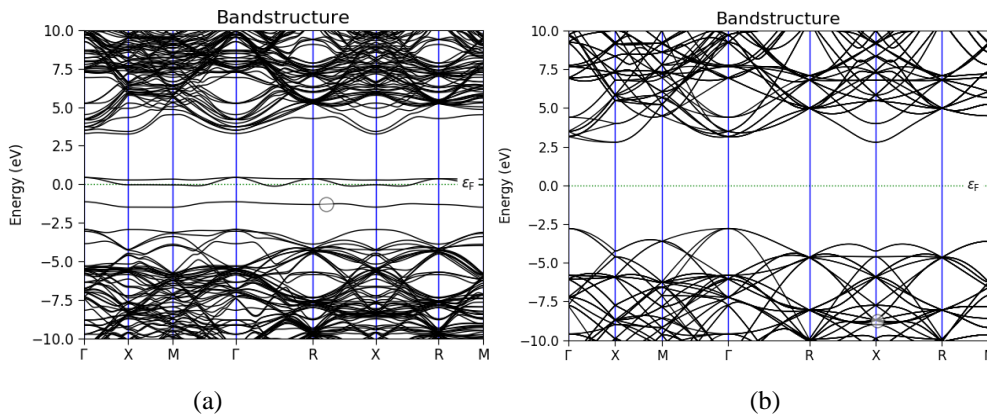

(a)

(b)

Figure 2. Calculated band structures for (a) defected diamond bulk by a pair of NV; (b) pure diamond bulk.

2. RESULTS AND ANALYSIS

Calculated band structures for bulk models are displayed in Figure 2. It shows that the bandgaps for the pristine diamond model are 5.57 eV (indirect) and 5.87 eV (direct), well matching to previously reported bandgap values [4]. There are three energy bands induced from the NV pair (Figure 2a), while the bandgap is a little wider (~6.3 eV). In the optical absorption coefficient results, there is a clear peak in the UV region (357 nm) for the NV defected diamond and a peak in the red region (729 nm) shown in Figure 3a. For the pristine model, there is only one peak in the wavelength of 142 nm (Figure 3b).
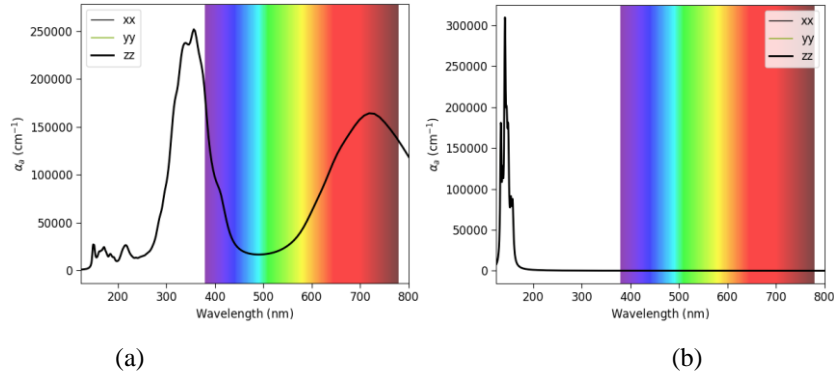

(a)　　　　　　　　　　　　　　　　　　(b)

Figure 3. Calculated optical absorption coefficients for (a) defected diamond bulk by a pair of NV; (b) pure diamond bulk.

In the thin film calculations, NV center is at the fifth layer from the top surface shown in the Figure 4a. The calculated band structure (Figure 4b) for the slab model is very similar the that of bulk model, showing a 5.48 eV, which is very close to the bandgap value (5.47 eV) in the reference [4]. Similar to the results in the bulk model, there are three extra bands introduced by the NV defect, which leads to a profound optical absorption peak in the far infrared region (2156 nm), shown in Figure 4c. There is also a very clear peak at 661 nm.
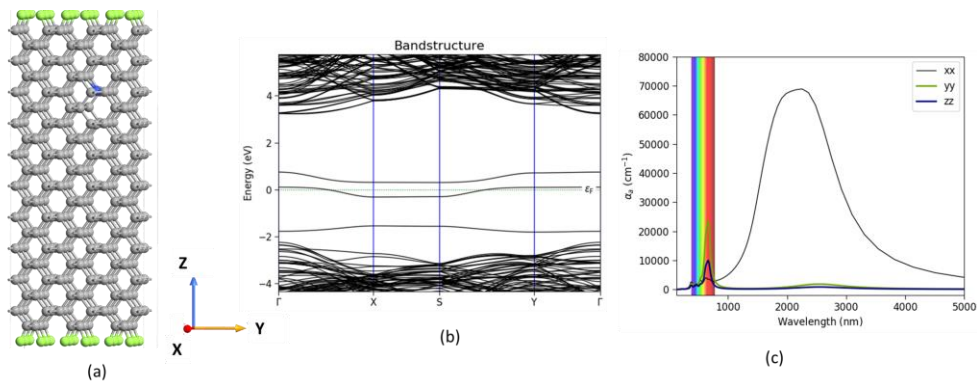

Figure 4. (a) Atomic structure of a NV center in diamond in the form of then film (20 layers of C terminated by Fluorene); (b) calculated band structure for the NV diamond in thin film; (c) optical absorption coefficient for the slab model.

More calculations were conducted for the localized electron difference density (EDD), which is the difference between the self-consistent valence charge density and the superposition of atomic valence densities. Figure 5 gives the EDD and electron difference potential (EDP) along the z-axis (thickness direction of the slab). Clearly seen from the Figure 5 that there is large electron potential on both surfaces, and there is a potential dip in the NV site (Figure 5b). EDD shows that there is large charge transfer on the surfaces, positive EDD indicates more electrons and negative EDD indicates less electrons.
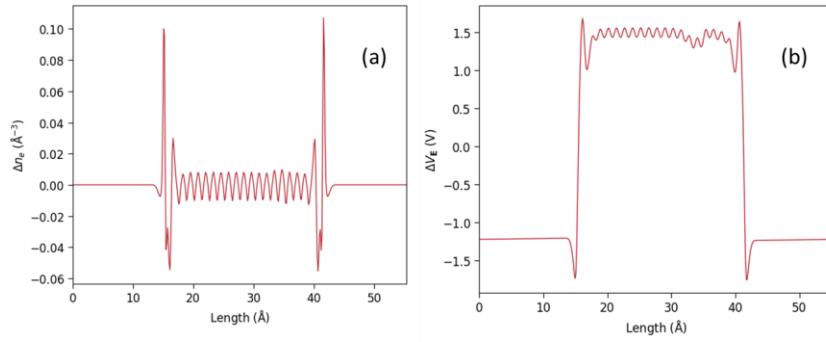
Figure 5. (a) The sum of electron difference density along the thickness direction of the diamond thin film; (b) The sum of electron difference potential along the thickness direction.

Further calculations were made to display iso-surfaces of the electron density obtained from the ground state DFT, shown in Figure 6. It unveils that the large electron densities are located around the atomic bonds, particularly around fluorene atoms on the surface. Three-dimensional DOS is shown in the left figure, while the right figure shows the 2D mapping.
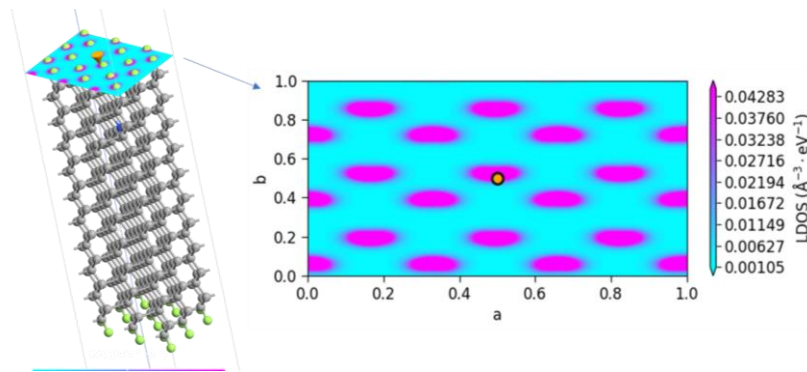

Figure 6. Local density of states of the NV diamond slab at the top surface.
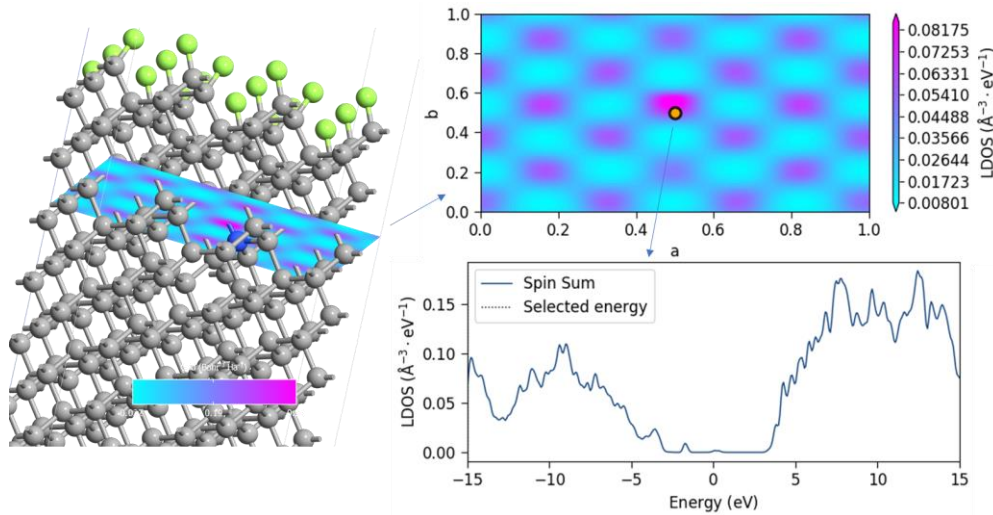

Figure 7. Local density of states at the cross-section of NV site.

Moving the iso-surface from the top surface to the location where the NV site is, it is obvious the electron densities are no longer periodically distributed. Instead, localized higher DOS around the NV center means that NV center is likely to be negatively charged, which agrees with the experiment reported previously [5].

3. CONCLUSION

To summarize, DFT simulation of the NV defect in diamond has been conducted for band structures, optical absorption spectrum, and electron density distributions. It is found that the slab model gives much closer bandgap values to the experimental value. There is a strong optical absorption peak in the far infrared region for slab model of the NV defected diamond. Electron density analysis shows that there is higher DOS around the NV site, creating electron traps, which underpins the fact that the NV centers are normally negatively charged. Future analysis will be conducted to take account of spin properties. The concept of using NV defected diamond as the sensor to detect the weak electromagnetic waves for neutrino mass determination offers much advantage to achieve higher signal-to-noise ratio as it is a purely optical (cold) detection mechanism.

REFERENCES

[1]   L. Childress, and R. Hanson, "Diamond NV centers for quantum computing and quantum networks," MRS Bulletin, 38(2), 134-138 (2013).

[2]   L. Li, "Single-layer Ga2O3/graphene heterogeneous structure with optical switching effect," Carbon Trends, 7, 100153 (2022).

[3]   W. Shen, G. Wu, L. Li et al., "Fluorine-terminated diamond (110) surfaces for nitrogen-vacancy quantum sensors," Carbon, 193, 17-25 (2022).

[4]   C. J. H. Wort, and R. S. Balmer, "Diamond as an electronic material," Materials Today, 11(1), 22-28 (2008).

[5]   K. Ohashi, T. Rosskopf, H. Watanabe et al., "Negatively Charged Nitrogen-Vacancy Centers in a 5 nm Thin 12C Diamond Film," Nano Letters, 13(10), 4733-4738 (2013).