A paramagnetic neutral $C_B V_N$ center in hexagonal boron nitride monolayer for spin qubit application

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Abstract

First-principles calculations in combination with group theory analyses were employed to study the spin-polarized electronic structures of $C_B V_N$ centers consisting of a nitrogen vacancy and a substitutional carbon atom in hexagonal boron nitride (*h*-BN) monolayer with different charge states. It is clarified that the paramagnetic neutral $C_B V_N$ center is stable in the *n*-type *h*-BN monolayer. The neutral defect center possesses a triplet ($S=1$) ground state and with two spin-conserved optical vertical transition. Its spin coherence time is estimated to be 3.9 ms at $T = 0$ K by a simple scheme combining the mean-field theory and the first-principles calculations. The results indicate that the neutral $C_B V_N$ center is very suitable for achieving spin qubit.

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1. Introduction

Recently, seeking potential materials with defects for use as qubit has attracted extensive attention. Among the possibilities in the solid state, the most successful defect is a negatively charged nitrogen-vacancy (NV^{-1}) center in diamond which consists of a carbon vacancy and an adjacent substitutional nitrogen impurity.[1-3] The center has many important features which have led to dramatic quantum effects, such as long coherence times,[4, 5] single spin readout[6] and spin polarization in moderate optical fields, *etc.*[6, 7] Particularly, it can be initialized, manipulated, and measured all at room temperature [8]. Therefore, the diamond NV^{-1} center has been proved to be a leading qubit candidate. [9-12]

Considering the difficulty in fabricating devices on diamond, it is very necessary to seek other qubit candidates which exhibit similar electronic and optical characteristics. Weber *et al.* have presented a list of physical criteria that a candidate deep center and its host should meet.[13] Following the criteria, various vacancy complexes in wide-band-gap semiconductors, such as a negatively charged $\text{N}_\text{C}\text{V}_\text{Si}^{-1}$ defect center in $4H\text{-SiC}$, [14-16] a neutral $\text{V}_\text{Ga}\text{O}_\text{N}$ center in GaN [17] and a neutral $\text{V}_\text{Al}\text{O}_\text{N}$ center in AlN *etc.*, [18] have been proposed to be promising candidates for quantum information applications.

So far, the proposed solid-state qubit candidates are all based on the tetrahedrally bonded bulk materials. In this work, we presented our efforts to seek the promising defect centers in layer semiconductor materials. We elucidated that a $\text{C}_\text{B}\text{V}_\text{N}$ center composing of a substitutional carbon atom at boron site (C_B) and adjacent nitrogen vacancy (V_N) in hexagonal boron nitride ($h\text{-BN}$) monolayer is very suitable for achieving spin qubit. Hexagonal boron nitride is an excellent wide band gap material with remarkable physical properties and chemical stability. As a structural

analogue of monolayer graphene, the monolayer h -BN has been fabricated experimentally.[19-21] Single atomic layer thickness makes it possible to fabricate devices with channels that are extremely thin. Additionally, the high thermal and chemical stability of h -BN makes this material suitable for a range of high-temperature and high-pressure applications. On the basis of density-functional theory (DFT) calculations and group theory (GT) analyses for the energetic stability and electronic structures of $C_B V_N$ centers in h -BN monolayer, it is revealed that a paramagnetic neutral $C_B V_N$ center is stable in the n -type h -BN monolayer and it possesses a triplet ($S=1$) ground state and a spin-conserved excited state with higher excitation energy. In addition, a simple scheme combining the mean-field theory and first-principles calculations were used to estimate the electron spin coherence time of the neutral $C_B V_N$ center. The obtained results indicate that the neutral $C_B V_N$ center is a promising candidate for spin coherent manipulation and qubit operation.

2. Calculation method

In order to study the energetic stability and electronic structure of the $C_B V_N$ center in different charge states, the spin-polarized density functional theory within the generalized gradient approximation (GGA) is adopted. As for the structural optimization and total energy calculations, the electron wavefunctions are expanded as linear combinations of pseudo atomic orbitals (LCPAOs),[22, 23] which are generated by using a confinement potential scheme with the cutoff radius of 5.0, 5.0, and 5.0 a.u. for B, N, and C, respectively .[24, 25] The electron-ion interactions are presented by the Troullier-Martins-type normconserving pseudopotentials with a partial core correction .[26] The electron-electron interaction was treated within the GGA in the form of Perdew-Burke-Ernzerhof (PBE) functional .[27] To simulate the defect centers, we employed a $7 \times 7 \times 1$ supercell consisting of

h-BN monolayer and the adjacent layers are separated by a vacuum region 15 Å. This supercell size ensures that defects are isolated from each other, avoiding the defect-defect interactions. In the structure relaxations, the atomic geometries are fully optimized until the Hellmann-Feynman forces are less than 1.0×10^{-3} hartree/a.u and the total energies are converged to 1.0×10^{-7} hartree with a Monkhorst-Pack *k*-point mesh of $5 \times 5 \times 1$. The calculated band gap of *h*-BN monolayer is about 4.6 eV.

3. Results and discussion

At first, we optimized the geometries of the $C_B V_N^q$ center in *h*-BN with different charge state *q* varying from +1 to -1. It is observed that the optimized atomic configurations of the $C_B V_N^q$ centers in the *h*-BN monolayer exhibit a perfect C_{2v} symmetry. For instance, Fig. 1 shows the optimized structure of the $C_B V_N^0$ center. The two B-C distances between the substitutional C and B atoms of the V_N are identical to 2.76 Å, while the B-B distance between the two B atoms adjacent to the V_N is 2.61 Å. The B-B distances are elongated by about 4% compared to the equilibrium B-B distance 2.51 Å in perfect *h*-BN. To determine the stability of the $C_B V_N$ center in different charge states, it is crucial to calculate the defect formation energies (E^f), which depend on the defect charge states (*q*) and the Fermi level (ϵ_F). [28] The following expression gives the formation energy of the $C_B V_N^q$ center in the *h*-BN monolayer:

$$E^f[\text{BN}: C_B V_N^q] = E_{tot}[\text{BN}: C_B V_N^q] - E_{tot}[\text{BN}: \text{monolayer}] - \mu_C + \mu_{BN} + q[\epsilon_F + \epsilon_{VBM} + \Delta V],$$

where $E_{tot}[\text{BN}: \text{monolayer}]$ and $E_{tot}[\text{BN}: C_B V_N^q]$ are the total energies of the perfect *h*-BN supercell and of the defective supercell containing the $C_B V_N^q$ center in the charge state *q*. μ_C is the chemical potential of an carbon atom and μ_{BN} is the energy per BN pair, which are calculated from

the graphene and a perfect *h*-BN monolayer, respectively. ε_F is the Fermi level reference to the valence-band maximum (VBM) in *h*-BN monolayer, ε_{VBM} . An alignment procedure (ΔV) is adopted to align the averaged electrostatic potentials between the defective supercell and the host.

The calculated formation energies of the $C_B V_N$ center in different charge states are plotted in Fig.

2. For a given ε_F , the formation energies in different charge states can be compared to determine which charge state is the most stable. It shows clearly that the stable charge state is sensitive to the position of Fermi level, where the $C_B V_N^0$ center is stable when ε_F is between 2.7 eV and 3.4 eV. The result implies that the $C_B V_N^0$ center exists mainly in the *n*-type *h*-BN monolayer. Furthermore, we can estimate the maximum concentration of the $C_B V_N^0$ center under thermal equilibrium by using the expression " $c = c_N \exp(-E^f / k_B T_m)$ ". Here, c_N is the atomic concentration of nitrogen in *h*-BN monolayer, $T_m = 3000$ °C is the melting point of *h*-BN monolayer, and k_B is the Boltzmann constant. Based on the calculated E^f , the equilibrium concentration of the $C_B V_N^0$ center is estimated to be 1.4×10^{-13} , which is so low that neither a direct magnetic coupling nor a long-range magnetic order of the defects is expected. Therefore, the $C_B V_N^0$ centers behave independently like paramagnetic centers.

Figure 3 presents the calculated density of states (DOS) of the $C_B V_N^q$ centers in the *h*-BN monolayer in the charge states $q = +1, 0$, and -1 . It is evident that the distinct defect levels are located in the band gap. These levels are relatively narrow, implying that the $C_B V_N$ centers possess the highly localized electronic bound states. The molecular orbitals (MOs) theory is applied to analyze the defect levels of the C_{2v} -symmetric $C_B V_N$ center in the *h*-BN monolayer. Generally, the single molecular orbitals of the $C_B V_N$ center can be constructed by using linear combinations of the orbitals of the atoms nearest to the vacancy. Therefore, the two sp^2 boron orbitals (σ_1 and σ_2) and one sp^2

carbon orbital (σ_3) can generate three sigma molecular orbitals. Moreover, the two boron p_π orbitals (π_1 and π_2) and one carbon p_π orbital (π_3) can form three pi molecular orbitals. The molecular orbitals of the $C_B V_N$ center are then constructed as the orthonormal, real linear combinations of the atomic orbitals by using the projection operator techniques, i.e., three sigma molecular orbitals and three pi molecular orbitals, respectively,

$$\begin{cases} a_1^\sigma = \alpha\sigma_3 + \sqrt{\frac{1-\alpha^2}{2}}(\sigma_1 + \sigma_2) \\ a_1^{\sigma*} = \sqrt{1-\alpha^2}\sigma_3 - \frac{\alpha}{\sqrt{2}}(\sigma_1 + \sigma_2) \\ b_2^{\sigma*} = \frac{1}{\sqrt{2}}(\sigma_1 - \sigma_2) \end{cases},$$

and

$$\begin{cases} b_1^\pi = \beta\pi_3 + \sqrt{\frac{1-\beta^2}{2}}(\pi_1 + \pi_2) \\ b_1^{\pi*} = \sqrt{1-\beta^2}\pi_3 - \frac{\beta}{\sqrt{2}}(\pi_1 + \pi_2) \\ a_2^{\pi*} = \frac{1}{\sqrt{2}}(\pi_1 - \pi_2) \end{cases}.$$

Here, $0 \leq \alpha, \beta \leq 1$ are parameters that determine the extent to which the carbon atomic orbital is mixed in the a_1^σ , $a_1^{\sigma*}$, b_1^π and $b_1^{\pi*}$ MOs. According to the C_{2v} point group, the a_1^σ and $a_1^{\sigma*}$ MOs transform as the totally symmetric irreducible representation (IR) A_1 , while the $b_2^{\sigma*}$ MO transforms as the function of the IR B_2 . On the contrary, the b_1^π , $b_1^{\pi*}$ and $a_2^{\pi*}$ MOs possess respectively B_1 , B_1 and A_2 symmetries. These defect electronic states align in the order of $E(a_1^\sigma) < E(a_1^{\sigma*}) < E(b_1^\pi) < E(b_1^{\pi*}) < E(b_2^{\sigma*}) < E(a_2^{\pi*})$.

The above MOs and group theory enable us to clarify the symmetries of the defect energy levels. In the case of the $C_B V_N^0$ center, there are four dangling-bond electrons occupying the above spin-polarized MOs. As shown in Fig. 3(b), the spin-polarized calculation generates three up-spin and two down-spin defect states inside the band gap. For the up-spin defect states, there are totally three occupied states below the Fermi level, one merged with the valence band possessing the A_1

symmetry and other two in the band gap transforming as A_1 and B_1 , respectively. In addition, there are three unoccupied states (one among them is below the conduction-band minimum (CBM) and two in the conduction band) possess the B_1 , B_2 , and A_2 symmetries, respectively. However, for the down-spin defect states, there is only one occupied state (transforms as A_1) below the valence-band maximum (VBM). The two unoccupied states in the band gap possess the A_1 and B_1 symmetries, respectively. This is in good agreement with the volumetric visualization of the electron density isosurfaces for each of the calculated defect levels. The isosurfaces of spin density corresponding to highest occupied molecular orbital (HOMO) (up-spin) and lowest unoccupied molecular orbital (LUMO) (down-spin) are presented in Fig. 4. Obviously, the HOMO and LUMO possess B_1 and A_1 symmetries, respectively. Moreover, the HOMO mainly arises from the carbon p_π orbital, whereas the LUMO contains contributions from both carbon sp^2 and two boron sp^2 orbitals. Consequently, the calculations have reproduced the expected symmetry and ordering of the single electron states. The ground state spin configuration of the $C_B V_N^0$ center is confirmed to be $a_1^\uparrow a_1^\downarrow a_1^\uparrow b_1^\uparrow$.

According to the discussions mentioned above, it can be easily draw the energy level diagram for the $C_B V_N^0$ center as shown in Fig. 5(b). Similarly, the defect energy level diagrams for other charge states based on the calculated DOS are obtained and shown together in Fig. 5. These diagrams show the defect-induced levels and their occupations in a particular charge state, from which the spin state can be deduced. Consequently, the net spin of the $C_B V_N^q$ center is obtained as $S = 1/2, 1$, and $1/2$ for the charge state $q = +1, 0$, and -1 , respectively. It is clear that the $C_B V_N^0$ center is of a net spin of $S = 1$, which is a spin triplet essential for qubit application.[13, 29]

The possibility of using neutral $C_B V_N$ centers to achieve spin qubit operation is discussed

subsequently. Our first-principles calculations indicate that the occupied b_1^π and empty $b_1^{\pi*}$ states of the neutral $C_B V_N$ center are well isolated from the bulk bands with spacing larger than 1.6 eV, as shown in Fig. 3(b). Such feature ensures thermal stabilities of the states of the center, which is important to maintain the spin information of the defects. Furthermore, the $b_1^{\pi*}$ acceptor level is far away enough to avoid the interference of the thermal excitation of the host electrons. From Fig. 5(b), it is observed that the $C_B V_N^0$ center has a triplet ($S=1$) ground state with electron configuration of $(a_1^\sigma)^2 a_1^{\sigma*} b_1^\pi$ and a spin-conserved triplet excited state with configuration of $(a_1^\sigma)^2 a_1^{\sigma*} b_1^{\pi*}$. Therefore, spin manipulation of the $C_B V_N^0$ center could be achieved via the transition between these ground and excited states.

Because the photon absorption or emission of a defect center will lead to simultaneous thermal motions of the surrounding atoms of the defect center (i.e., the Franck-Condon principle), it is necessary to calculate the configuration-coordinate diagram of the $C_B V_N^0$ center.[30, 31] For example, the energy of vertical absorption transition could be calculated using the total energy of the excited state and that of the ground state within DFT, where the energy of the excited state was calculated using the ground state structure configuration but with a constrain occupation by promoting one electron from the $b_1^{\pi\uparrow}$ state to $b_1^{\pi*\uparrow}$ state. Similarly, the stable of excited state obtained by relaxed the structure with promoting one electron in $b_1^{\pi*\uparrow}$ state and the energy of the vertical emission transition was calculated using the excited state structural occupation. According this method, we changed successively the coordinates of the surrounding atoms along their relaxation directions and calculated the energies of the defect levels associated with the electron transition from the ground state to the excited state by keeping the C_{2v} symmetry fixed. In order to accurately calculate the zero-phonon-line (ZPL), the Heyd-Scuseria-Ernzerhof (HSE06) hybrid functional is

adopted.[32, 33] Apparently, the HSE06 functional improves not just the band gap of perfect semiconductor but also the internal defect transition energy as well. The results are illustrated in Fig. 6. It can be found that, for the spin-up electrons, the photon absorption and emission of the vertical transitions between $b_1^{\pi\uparrow}$ and $b_1^{\pi*\uparrow}$ levels are 1.71 and 1.40 eV, respectively, while the ZPL and the Franck-Condon shift are respectively 1.61 and 0.21 eV. However, the energies of the absorption, emission, and ZPL photons are 2.91, 2.09 and 2.23 eV, respectively, for the transitions between $a_1^{\sigma*\uparrow}$ and $b_1^{\pi*\uparrow}$ levels. The calculated ZPL of 1.61 eV is well consistent with the experiment observed value of GC-2 (1.63 eV) in BN.[34] These results indicate that the $C_B V_N^0$ center in the *h*-BN monolayer provides more degrees of freedom for the spin manipulation.

Taking into account that the electron spin coherence time of the $C_B V_N^0$ center is very important for the spin coherent manipulation and qubit operation, the hyperfine interaction of the defect-electron spin and the surrounding nuclear spins are estimated by combining the mean-field theory and first-principles calculations, through the Fermi contact interaction[18, 35]

$$\Delta E_{hyp} = \sum_n A_H \Delta N_n \bar{I}_n s .$$

Here, A_H is the hyperfine coupling constant, ΔN_n is the fraction of the defect electron being of the Fermi contact interaction with the n th nucleus, $\bar{I}_n = \sum_i \alpha^i I_n^i$ is the average value of spin of the n th nucleus in the supercell, where α^i and I_n^i are respectively the natural abundance and nuclear spin of the i th isotopes of the n th nucleus, and $s = 1/2$ is the value of the defect electron spin. According to the uncertainty principle, the electron spin coherence time at $T = 0$ can be estimated as $\Delta\tau = \hbar/2\Delta E_{hyp}$ with $\hbar = 6.5822 \times 10^{-16}$ eV being the Planck's constant. Based on the calculated LCPAO coefficients and the primary atomic orbitals of B 2s, N 2s, and C 2s, we get the electron-nucleus spin interaction $\Delta E_{hyp} = 8.345 \times 10^{-14}$ eV and the spin coherence time $\Delta\tau = 3.9$ ms at $T = 0$ for the

$C_B V_N^0$ center. It is noteworthy that $\Delta\tau$ will decrease considerably with increasing temperature. For instance, $\Delta\tau$ of the diamond NV^{-1} center is about a few seconds at low temperature but only 1.8 ms at room temperature.[9, 36-38] Therefore, it is expected that $\Delta\tau$ of the $C_B V_N^0$ center is in the order of μs at room temperature. This implies that the spin coherence time of the $C_B V_N^0$ center is suitable for qubit operation.

4. Conclusions

In summary, the energetic stability and electronic structure of the $C_B V_N$ center in the h -BN monolayer with different charge states are investigated by first-principles calculations. By combining the calculations, the molecular orbital theory and the group theory, it is elucidated that the neutral $C_B V_N$ center is paramagnetic and stable in the n -type h -BN monolayer. In addition, such defect center possesses a triplet ($S = 1$) ground state and a spin-conserved excited state. Its spin coherence time is in the order of ms at $T = 0$. These results indicate that the neutral $C_B V_N$ center is a promising candidate for spin coherent manipulation and qubit operation. This work opens a new avenue for the use of h -BN monolayer in quantum information and communication.

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Figure captions:

FIG. 1. The schematic structure of the neutral $C_B V_N$ center in the h -BN monolayer. The dashed lines indicate the C-B and B-B distances.

FIG. 2. Calculated formation energies (E^f) of $C_B V_N$ centers in different charge states as a function of Fermi levels (ε_F) in the h -BN monolayer.

FIG. 3. Calculated density of states (DOS) for $C_B V_N^q$ centers with different charge states in the h -BN monolayer, (a) $q = +1$, (b) $q = 0$, (c) $q = -1$. The energies are relative to the calculated chemical potentials ($E = 0$ eV) denoted by thin vertical lines.

FIG. 4. The spin-density isosurfaces of the highest occupied molecular orbital (HOMO) (a) and the lowest unoccupied molecular orbital (LUMO) (b) for the neutral $C_B V_N$ center in the h -BN monolayer. The boron and nitrogen atoms are colored in gray and blue, respectively.

FIG. 5. Schematic diagrams of defect energy levels of $C_B V_N^q$ centers with different charge states in the h -BN monolayer, (a) $q = +1$ and $S = 1/2$, (b) $q = 0$ and $S = 1$, (c) $q = -1$ and $S = 1/2$. Majority (minority) spins are denoted by upward (downward) arrows.

FIG. 6. Calculated configuration-coordinate diagrams of spin-conserved optical vertical transition for the $C_B V_N^0$ center in the h -BN monolayer. Absorption, emission, and ZPL transitions are indicated along with their energies in (eV).

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Fig. 2. G.D. Cheng et al.

Fig. 3. G.D. Cheng et al.

Fig. 4. G.D. Cheng et al.

Fig. 5. G.D. Cheng et al.

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