



Full length article

# Theory of sulfur-vacancy defect in diamond: a comparable $NV^{-1}$ isoelectronic center

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## ABSTRACT

A color center in diamond which is a comparable  $NV^{-1}$  isoelectronic center is predicted based on first-principles electronic structure calculations. The defect consists of a substitutional sulfur and an adjacent carbon vacancy (S-V). We find that the S-V center is optically accessible with two zero-phonon line of about 1.12 and 1.22 eV. The S-V center also shares many of the characteristics of the  $NV^{-1}$  center in diamond. A prominent spin coherence time is predicted by combining first-principles calculations and a mean-field theory for spin hyperfine interaction, and is at the same level with that of  $NV^{-1}$  center in diamond. Furthermore, the neutral S-V center in diamond provides more degrees of freedom for spin manipulation than the  $NV^{-1}$  center in diamond.

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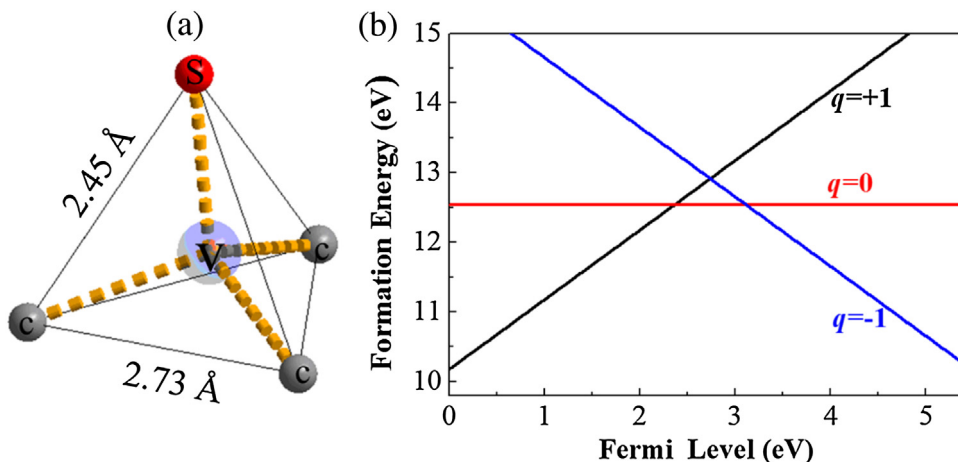
## 1. Introduction

Spin carrying defect levels in solids, which can be optically manipulated, have recently become versatile system for qubits application [1–4]. Recently, the negatively charged state nitrogen-vacancy ( $NV^{-1}$ ) center has emerged as promising candidates for a number of applications, ranging from high spatial resolution imaging to quantum computation [5,6]. At low temperatures, the optical transitions of  $NV^{-1}$  center become very narrow and can be coherently manipulated, allowing for spin-photon entanglement generation for quantum communication and all optical control [4,7].

Considering the  $NV^{-1}$  center emission bandwidth of about 100 nm at room temperature, which cause the emission of single photons in the ZPL extremely weak. [8]. Thereby, other defect center that behaves like the diamond  $NV^{-1}$  center would be greatly valuable. Because a neutral sulfur-vacancy (S-V) center composing of a substitutional sulfur atom at the carbon site and an adjacent carbon vacancy in the diamond is structurally analogous to and isoelectronic with  $NV^{-1}$  center, it was natural to consider the diamond SV center as a plausible qubit candidate. Indeed, the sulfur impurities play an important role in diamond growth and the sulfur-related defects in diamond have been studied both theoretically and experimentally [9–13]. In this work, spin-polarized defect energy levels, defect formation energies, spin-conserved optical transition, and spin coherence time of S-V centers in diamond were studied by comprehensive theoretical calculation. The obtain results revealed that the neutral S-V center was stable in both of the *n* and *p*-type diamond and possesses an *S* = 1 triplet ground state and two spin-conserved excited states with two zero-phonon lines (ZPLs). Moreover, the spin coherence time of the neutral

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**Fig. 1.** (a) Optimized structure of the neutral S-V center in diamond. Dash lines indicate the orientations of the dangling bonds. Blue, red, and gray balls indicated carbon vacancy, sulfur, and carbon atoms, respectively. (b) Calculated formation energies ( $E^f$ ) of the S-V center in different charge states as a function of the Fermi level ( $\varepsilon_F$ ) in diamond. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

S-V center was estimated by using a simple scheme combining a mean-field theory and first-principles calculations. All these indicate that the neutral S-V center was a promising candidate for spin coherent manipulation and qubit application.

## 2. Calculation method

The first-principles calculations in the framework of density functional theory (DFT) and norm-conserving pseudopotential method were performed [14,15]. The generalized gradient approximation (GGA) proposed by Perdew, Burke, and Ernzerhof (PBE) was adopted [16]. The basic functions consist of pseudoatomic orbitals are specified as  $s^2p^2d^1$  for both the carbon and sulfur atoms. For the structural optimization and total energy calculations, the electron wavefunctions are expanded as linear combinations of pseudoatomic orbitals (LCPAOs) [17,18], with the cutoff radius of the orbitals are 5.0 and 7.0 a.u. for carbon and sulfur atoms, respectively [19]. The real-space grid technique was used with the energy cutoff of 200 Ry in numerical integrations of Kohn-Sham equation and the Poisson equation is solved with the fast Fourier-transformation algorithm. To model the defect centers, a cubic supercell containing 63 atoms and a single carbon vacancy were employed. The Brillouin zone (BZ) integration is carried out by using a  $4 \times 4 \times 4$  Monkhorst-Pack special  $k$ -point mesh. In the structure relaxations, the atomic geometries were fully optimized until the Hellmann-Feynman forces were less than  $1.0 \times 10^{-4}$  Hartree/a.u. and the total energies were converged to  $1.0 \times 10^{-7}$  Hartree.

## 3. Results and discussion

The optimized configuration of a neutral S-V center in diamond is shown in Fig. 1(a). The three S-C distances between the substitutional S atom and the C atoms nearest to the vacancy are identical to 2.45 Å, while the C-C distances between the C atoms adjacent to the vacancy are 2.73 Å. Thereby, the neutral S-V center possesses a perfect  $C_{3v}$  symmetry [20,21]. The same symmetric property is also observed in the other charged state  $(S-V)^q$  centers. To determine the stability of the S-V center in different charge states (especially the stability of the neutral S-V center promising for qubit applications), it is crucial to calculate the defect formation energies ( $E^f$ ), which depend on the defect charge states ( $q$ ) and Fermi level ( $\varepsilon_F$ ). The  $E^f$  can be extracted from the total energies via first-principle calculations as [22,23]

$$E^f [C : (S-V)^q] = E_{tot} [C : (S-V)^q] - E_{tot} [C : bulk] - \mu_S + 2\mu_C + q (\varepsilon_F + E_{VBM}^{bulk} + \Delta V).$$

Where  $E_{tot} [C : (S-V)^q]$  and  $E_{tot} [C : bulk]$  are the total energies of the perfect diamond supercell and of the defective supercell containing the  $(S-V)^q$  center in the charged state  $q$ .  $\mu_S$  and  $\mu_C$  are chemical potentials of S and C atoms in the sulfur crystal and in the diamond crystal.  $\varepsilon_F$  is the Fermi level reference to the valence band maximum (VBM) of the diamond crystal ( $E_{VBM}^{bulk}$ ). An alignment procedure ( $\Delta V$ ) is adopted to align the averaged electrostatic potentials between the defective supercell and the host. The calculated formation energies as a function of  $\varepsilon_F$  are plotted in Fig. 1(b), where each line represents the formation energy of the defect in a certain charged state  $q$ . For  $\varepsilon_F$  between 2.14 eV and 3.33 eV, the neutral S-V center is stable. This implies that the neutral S-V centers exist in the both of  $n$  and  $p$ -type diamond.

Fig. 2 compares the electronic structures of an  $NV^{-1}$  and  $SV^0$  center calculated using a 63-atoms supercell. Similar to the  $NV^{-1}$  center in diamond, the  $SV^0$  center also introduces optically active defect states, i.e., occupied  $u$  and unoccupied  $v$  and  $e_{xy}$  minority-spin states (shown in blue in the figure), deep inside the band gap, thereby, enabling optical probe and/or

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