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Defects in SiC for Quantum Computing

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Abstract: Many novel materials are being actively considered for quantum information science and for realizing high-performance qubit operation at room temperature. It is known that deep defects in wide-band gap semiconductors can have spin states and long coherence times suitable for qubit operation. We theoretically investigate from ab-initio density functional theory (DFT) that the defect states in the hexagonal silicon carbide (4H-SiC) are potential qubit materials. The DFT supercell calculations were performed with the local-orbital and pseudopotential methods including hybrid exchange-correlation functionals. Di-vacancies in SiC supercells yielded defect levels in the gap consisting of closely spaced doublet just above the valence band edge, and higher levels in the band gap. The divacancy with a spin state of 1 is charge neutral. The divacancy is characterized by C-dangling bonds and a Si-dangling bonds. Jahn-teller distortions and formation energies as a function of the Fermi level and single photon interactions with these defect levels will be discussed. In contrast, the anti-site defects where C, Si are interchanged have high formation energies of 5.4 eV and have just a single shallow defect level close to the valence band edge, with no spin. We will compare results including the defect levels from both the electronic structure approaches.

INTRODUCTION:

The field of quantum information science has been rapidly expanding exploring new materials suitable for realizing quantum bits (qubits). [1] Qubits are the basic construct of quantum information and consist of a spin in a solid that is largely decoupled from its environment. The qubit is an intrinsically quantum object, that can be utilized to store and process information. [1] Many materials have been considered for realizing qubits. Wide band gap semiconductors are particularly attractive for qubits [1,2,3], since they are well known to have point defects, that have deep defect levels within the band gap. Many of this point defects can have high spin states depending on the position of the Fermi level. Moreover, since these defect levels are separated in energy from the band edges in wide band gap semiconductors, the electronic states of these defects are well localized and are not appreciably extended into the host semiconductor. Such isolated high spin states in

wide band gap semiconductors have great potential for qubit operation since they are expected to have long coherence times, an essential requirement for quantum computing.

Building upon the understanding of nitrogen vacancy (NV) center in diamond allows us to identify similar defects in other systems. The properties of vacancies and NV defects in diamond and 4H-SiC are comparable. [3] The ranges of the Fermi level that produce the appropriate defect spin and charge states also allow for spin-conserving excitations. These excitations can be represented in terms of defect-level diagrams where the vacancy center in SiC is very similar to that of the defect center in diamond. [3] However, the splitting between the levels in SiC is much smaller than those in diamond, primarily due to the reduced dangling bond overlap caused by the crystal's larger bond lengths. [3] The isolated silicon vacancies in SiC can possess properties similar to those of the NV centers and function as qubits.

APPROACH

We have chosen the prototypical system of silicon carbide for illustration of defects for quantum information science. Although the nitrogen-vacancy (NV) center is very well studied in diamond, the intrinsic defects in silicon carbide are an alternate system with great potential. Ab-initio density functional simulations were performed on 4H-silicon carbide (4H-SiC) to investigate the nature of defects and their potential for qubits. We utilized a 96 atom supercell of 4H-SiC, consisting of 3x4x1 primitive unit cells employing the ABAC stacking sequence, similar to other DFT simulations. [1-3]

RESULTS

We describe results for two different simulation approaches utilizing local orbital and plane wave-based density functional approaches, to contrast the relative merits of each approach. The former one is SIESTA [4] whose efficiency stems from the use of strictly localized basis sets and from the implementation of linear-scaling algorithms which can be applied to much larger systems that in plane-wave based approaches. A very important feature of the code is that its accuracy can be tuned in a wide range, from quick exploratory calculations to highly accurate simulations matching the quality of other approaches, such as plane-wave and all-electron methods. The latter is Vienna Ab initio Simulation Package (VASP), which is suitable for atomic scale electronic structure calculations and quantum-mechanical molecular dynamics from first principles.

SIESTA computations

In the first approach, the simulations utilized the local orbital method of SIESTA (Spanish Initiative for Electronic Simulations with Thousands of Atoms), with norm-conserving pseudopotentials for Si and C. The Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional based on the general gradient approximations (GGA) was used to treat the nonlocal exchange and correlation energies.

The relaxed 4H-SiC (in the 96-atom supercell) yielded a ground state energy of -134.5915 eV/atom, and Si-C bond lengths of (1.85 Å). The band gap was 1.88 eV, lower than the experimental value of 3.1 eV due to well-known problems with DFT-GGA simulations. We created a divacancy, by removing a pair of (Si,C) atoms, and relaxed the structure, which had an energy of 0.0796 eV/atom above the ground state (Fig. 1). This yields energy levels within the band gap shown in Fig. 2. There is a level at the top of the valence band followed by a doublet ~1 eV above the valence band edge and higher states.

For comparison with previous work [3], we have raised the conduction band edge to yield the correct band gap (Fig. 2). In the (111) layer in which the Si is removed, there remain three C-dangling bonds that contribute to the gap states. In the layer in which the C is absent, two Si atoms form a weak Si-Si bond of 2.82 Å, and in addition there remains a single Si dangling bond The neutral SiC defect has spin states filled according to Hund's rules, showing a total spin of S=1 (3 spins up; 1 spin down), for Fermi level positions near midgap (Fig. 2).

A natural occurring defect in semiconductors is the anti-site defect consisting of an interchange of a Si, C atom. It is of interest to examine the energetics of such anti-site defects since they can compete with and hinder the qubit operation. We examined the energetics of this anti-site defect by performing relaxation in the 96 atom supercell. This yields high energy of 5.38 eV above the ground state due to very strained and compressed Si-Si bond lengths of 2.175 Å (in one layer) accompanied by stretched C-C bond lengths of 1.60-1.63 Å in the adjoining (111) layer. It is unlikely that the antisite defect will affect the properties of the qubit in SiC structures.

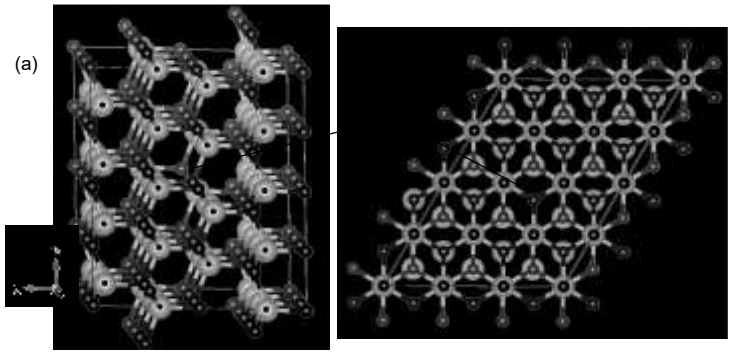

Figure 1. (a) 3D view, (b) 2D view of atomic structure of divacancy in 96 atom SiC supercell (Si and C are in Grey and black colors, respectively).

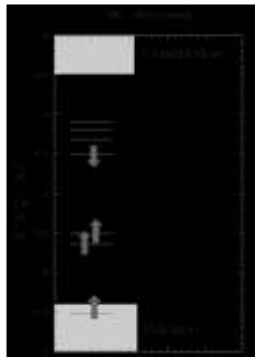

Figure 2. Energy levels of divacancy in 4H- SiC from Siesta calculations.

VASP calculations

In the second approach, we used the plane wave pseudopotential DFT total energy calculations based on the plane wave expansion within the Vienna Ab-initio Simulation Package (VASP). Here the one-electron orbitals, the electronic charge density,

and the local potential are expressed in plane wave basis sets. The interactions between the electrons and ions are described using the projector-augmented-wave (PAW) method. To alleviate the bandgap problem we employed generalised gradient approximation (GGA) as an exchange-correlation functional using the Perdew-Burke-Ernzerhof (PBE) scheme. The convergence criterion for the self-consistent calculations is 10^{-7} eV for the total energy per supercell cell, the Brillouin zone integration was performed on a 3×3×2 k-point grid using the tetrahedron method. We have used an energy cut-off of 500 eV for the electronic wave functions. Calculations were performed for a 96 atom 4H-SiC supercell and the 4H-SiC supercell with a relaxed divacancy defect. The electronic energy bands across the first Brillouin zone for the 2 cases are shown in Fig. 2. We obtained a band gap of ~2.2 eV (Fig. 3a) for the crystalline 4H-SiC with PBE approach- however, the value increases to 3 eV with the hybrid functionals (not shown here). The latter is in reasonable agreement with the experimental value of 3.17 eV. It should be noted, that when the PBE approximation was used the band gap reduced to 2.2 eV, close to the SIESTA calculation. The relaxed divacancy resulted in four flat defect bands within the band gap of the SiC, showing little variation in the energies across the Brillouin zone (Fig. 3b), indicating weak coupling with the host lattice.

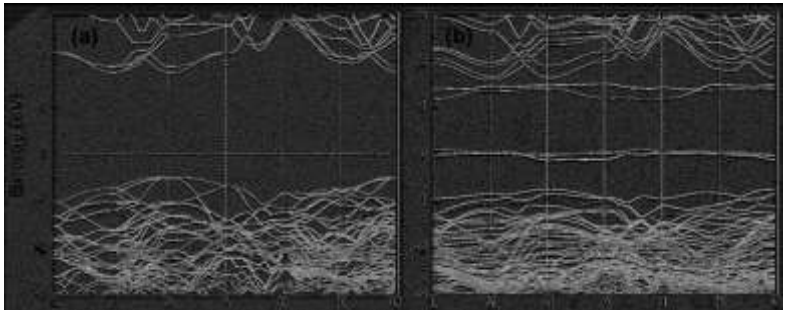

Figure 3. a) Electronic energy bands for crystalline 4H-SiC in a 96-atom supercell computed with the plane wave based VASP scheme. b) Electronic energy bands for the divacancy in 4H-SiC computed with the VASP scheme.

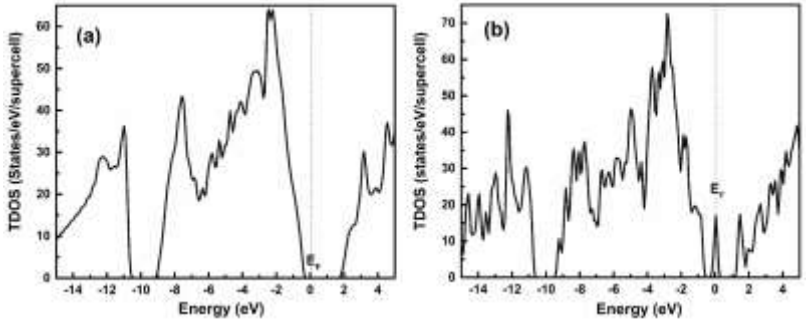

Figure 4. Electronic densities of states for 4H-SiC for a) the crystalline 4H-SiC and b) the divacancy in 4H-SiC computed with the plane wave based VASP scheme.

The electronic density of states (Fig. 4a) illustrates the energy gap of ~2.2 eV for 4H-Si, and the peak in the valence band DOS arising mostly from p-states. The DOS for the divacancy shows the defect bands within the band gap with minor changes in the valence band DOS. It is of interest to examine the individual defect levels in 4H-SiC by

plotting their values at the Gamma-point since these are needed for effective qubit operation. We show these results in Fig. 5

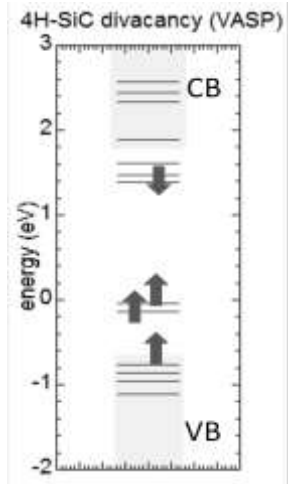

Figure 5. Energy levels with spin states for divacancies in 4H-SiC based on VASP computations

DISCUSSION

The world of defect centers is vast, and only one small subset of NV centers in diamond and divacancies in SiC have been studied so far in relation to quantum information [5]. A substantial research effort is necessary to determine which other classes of defect centers are compatible with the defect and host criteria. Many isolated substitutional or interstitial impurities may act as defect centers but no such center has been clearly identified. In the octahedrally coordinated hosts, optical spin polarization has been reported in vacancy-related complexes [5], but more exploration is required to determine what other features of these centers have in common with the NV center in diamond.

We have shown total energy calculations for divacancy states in 4H-SiC. Since SiC is a non-magnetic material, spin-polarized calculations are not necessary. However it is necessary to constrain the spin state by singly occupying defect states, and do further structural relaxation to obtain the energetics of the defects more precisely.

CONCLUSION

In summary, we have examined the divacancy in SiC for utilization as a high spin qubit state using theoretical ab-initio density functional calculations. We have utilized the local orbital based SIESTA method and compare it with the plane wave based VASP method. Our approach compares the strengths and shortcomings of both computational approaches in describing qubits in wide band gap semiconductors. We demonstrate that silicon carbide can sustain divacancies with spin 1 which are well suited for qubit operation. Other wide band semiconductors (e.g. GaN) also have considerable potential for high spin defect states that can act as qubits.

ACKNOWLEDGEMENTS

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