Investigation of quantum bits and single photon sources in silicon carbide by means of quantum mechanical simulations

PhD Thesis Booklet

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Introduction

Quantum description of elementary particles has induced a quest for exploiting their properties beyond the boundaries of classical physics. Measuring and controlling the quantum state of a system have implied a broad set of scientific results. Consequently, quantum technology has been grown to one of the most prosperous field of applied sciences.

Paramagnetic point defects embedded in wide-band-gap semiconductors may serve as single photon emitters and quantum bits (or qubits) being essential building blocks in realization of quantum communication and cryptography [Weber et al., 2010]. The most investigated member of this family so far has been the nitrogen-vacancy (NV) defect in diamond [Doherty et al., 2013]. Particularly, the qubit state is realized by the single negative charge state of the NV defect (NV^-) designated with the notion of NV center. The electronic structure exhibits S=1 spintriplet ground state with $m_S = 0, \pm 1$ spin projection quantum number and can be subject to the so-called optical spin polarization promoting the system into the $m_S = 0$ state. This procedure gives rise to the spin state to be optically initialized and read-out making NV center a desirable candidate for employing in quantum information processing.

Difficult processability of diamond in the semiconductor industry has induced a quest for other technologically mature materials which are capable of incorporating point defects exhibiting superior properties. Silicon-carbide (SiC) has proven to be a competitive candidate for this role with a wafer scale fabrication procedure and straightforward integrability to recent semiconductor devices.

Owing to the outstanding properties of NV center and the closely related lattice structure of SiC and diamond it is reasonable to investigate the NV center in SiC with respect to its physical properties, conditions of its formation, fabrication and application. Experimental identification of NV center in the 3C, 4H and 6H polytypes of SiC has been reported in preliminary photoluminescence (PL) and electron paramagnetic resonance (EPR) studies [von Bardeleben et al., 2015, von Bardeleben et al., 2016, Zargaleh et al., 2016, Zargaleh et al., 2018a, Zargaleh et al., 2018b]. However, local symmetry conditions of the corresponding SiC lattice structures give rise to a set of NV center configurations. In particular, NV center forms 1 defect configuration in the 3C, 4 configurations in the 4H and 6 different configurations in the 6H polytype. Although presence of NV center was confirmed by the experiments in the afore-mentioned polytypes, different defect configurations had not been fully resolved which is an inevitable step towards quantum technology applications.

According to previous experimental results, neutral divacancy in 4H SiC also provides

a suitable platform for the optical spinpolarization process [Christle et al., 2017]. However, emission of neutral divacancy appears only at above a — higher than expected — threshold energy as pointed out by PL and PL excitation (PLE) measurements [Wolfowicz et al., 2017], [Golter and Lai, 2017]. In the literature two explanations had been showed up both claiming that the phenomenon can be explained by switching the neutral qubit state into another charge state optically. This charge state can be back ionized (or reionized) to the neutral qubit state at a shorter wavelength photo-excitation than applied for the optical readout of the divacancy qubit. Nonetheless, their suggestions in terms of the 'dark' state are contradictory: one proposed the positive charge state divacancy [Golter and Lai, 2017], while the other argues the single negative charge state for this role [Wolfowicz et al., 2017].

Another drawback of NV center in diamond is that its emission falls far from the window being used in present-day telecommunication wavelength region. As reported by preliminary theoretical and experimental studies [Baur et al., 1997] transition metal point defects in SiC exhibit emission spectra close to the near-infrared (NIR) region and hence may be suitable for realization of single-photon emitters as building blocks for quantum communication. In particular, zero-phonon lines (ZPLs) related to vanadium defects in 6H SiC fall into the O-band (1260-1360 nm) of the telecom wavelength region in agreement with previous magnetic circular dichroism (MCD) and EPR studies [Kaufmann et al., 1997], [Kunzer et al., 1993]. In these studies the corresponding electronic structure were determined in a semi-empirical way employing crystal field theory, notwithstanding, *ab initio* calculations were not included. Similar experimental results had not yet been available in the literature for Vanadium impurity in 4H SiC, however, it is expected to be as unique as its counterpart in 6H SiC owing to the closely related lattice structure of 4H and 6H polytypes.

Objectives

Main target of my PhD work was to investigate point defects incorporated by SiC and reveal the conditions of employing them as qubit and/or single photon emitters.

My first objective was to support a theoretical model established for proposing a possible microscopic scenario responsible for the quenching mechanism in the PL spectrum of divacancy in 4H SiC. To this end, I calculated the corresponding charge transition levels for all divacancy configurations. The afore-mentioned model claims that the presence of divacancy in its negative charge state is responsible for the quenching phenomenon, whereas application as quantum bit requires the divacancy to be in the neutral charge state. Conse-

quently, charge state control of the neutral divacancy has to be ensured during application. I calculated temperature dependent photoionization spectra of the negative charge state of all divacancy configurations in order to find an excitation frequency with which optical charge state control can be realized.

My second goal was the unambiguous identification of NV center configurations in the most common polytypes of SiC along with the investigation of defect formation and application aspects. To this end, I determined the electronic structure, the positions of the ZPLs and the magnetic properties employing Density Functional Theory (DFT) calculations. In order to predict conditions of fabrication and application procedures, I calculated the concentration of NV centers in the different polytypes along with that of other native defects created by usual fabrication processes as chemical vapor deposition (CVD) and implantation.

The third aim of my work was to explain the PL spectrum originating from substitutional vanadium defect in 4H SiC. Since *ab initio* results are not available in the literature, I calculated the one-electron structures for both the corresponding ground and excited states. As a result I obtained the corresponding ZPLs and ground state spin-orbit splittings. Furthermore, I identified the sharp peaks in the corresponding PL spectrum accounting for the phonon-assisted optical transitions.

Methodology

During my PhD course I have carried out electronic structure calculations for point defects by means of Kohn-Sham (KS) DFT. Particularly, I employed the so-called PBE [Perdew et al., 1996] and HSE06 [Heyd et al., 2003] exchange-correlation functionals. For modelling the point defects I utilized 576 atom supercell for the 4H, 432 atom supercell for the 6H and 512 atom supercell for the 3C polytype. For the supercell method it is natural choice to expand the KS wavefunctions in plane wave basis set. In most cases, I used Γ-centered **K**-point meshes for sampling the Brillouin-zone. Only valence electrons were treated explicitely during the calculations, while core electrons were taken into consideration via the Projector Augmented Wave (PAW) [Blöchl, 1994] method. For electrically charged defects I employed the Freysoldt charge correction scheme [Freysoldt et al., 2009] on the total energy. As for the formation energies and neutrality equation for the considered defects in solids I used an already available code that I modified to suit my particular systems. I obtained PL spectra by using the Huang-Rhys (HR) approximation [Alkauskas et al.,

2014]. An already built code was available for this task as well, however, only for cubic supercells. I modified that code in order to be capable of obtaining HR spectra for point defects embedded in hexagonal environment.

New scientific results

1. Photoluminescence quenching of divacancy in 4H SiC.

Quenching of the neutral divacancy-related ZPLs in the corresponding PL spectra in 4H SiC has been reported in earlier studies [Wolfowicz et al., 2017,Golter and Lai, 2017,Magnusson et al., 2018]. Divacancy in 4H SiC forms 4 different defect configurations, namely *hh*, *kk*, *hk*, *kh* configurations, where the first letter denotes the site of Si vacancy (V_{Si}) while the second stands for the site of carbon vacancy (V_C) as *h* - hexagonal and *k* - cubic. All the four corresponding ZPLs exhibit the quenching mechanism. Recovery of the ZPLs is observed under illumination with excitation energies (threshold energies) of 1.310 eV (*hh* and *kk*), 1.321 eV (*hk*) and 1.281 eV (*kh*) [Magnusson et al., 2018].

Two competing models are available in the literature explaining the quenching mechanism. Both models assume that $V_{Si}V_C$ is not present in its neutral charge state (qubit state) resulting in the absence of the corresponding ZPLs. The recovery process occurs after the reionization of $V_{Si}V_C$ to its neutral charge state and hence the reported threshold energies correspond to the reionization energies from the so-called dark state to $V_{Si}V_C^0$. However, identification of the dark state is contradictory: in Ref. [Golter and Lai, 2017] the positive charge state [$V_{Si}V_C^+$] is proposed, while in Ref. [Wolfowicz et al., 2017] the dark state is identified as the negative charge state [$V_{Si}V_C^-$].

I studied the quenching mechanism at different temperatures by DFT calculations. For modeling the lattice structure I utilized 576 atom supercell along with HSE06 hybrid functional to describe the electronic structure. Reionization process of $V_{Si}V_C^+ \rightarrow V_{Si}V_C^0$ can be carried out by promoting an electron from the valence band to the ingap *e* state, while by electron transition from the *e* state to the conduction band is needed for the $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ process. In this way, energy required for the $V_{Si}V_C^+ \rightarrow V_{Si}V_C^0$ reionization process can be approximated as the energy difference between the valence band edge and the (+/0) charge transition level, while that for the $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ process can be estimated by the energy gap between the (0/-) charge transition level and the conduction band edge. I report my results in the following 2 thesis points.

(a) **Quenching of divacancy-related features in the PL spectrum at T $\approx$ 0 K.**

For energies of $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ transition I obtained 1.245 eV (hh), 1.209 eV (kk), 1.174 eV (hk) and 1.307 eV (kh). For the $V_{Si}V_C^+ \rightarrow V_{Si}V_C^0$ process I obtained 1.070 eV (hh), 1.010 eV (kk), 1.051 eV (hk) and 1.081 eV (kh).

Calculated values for the $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ transition show good agreement with the experimental threshold energies in contrast those for the $V_{Si}V_C^+ \rightarrow V_{Si}V_C^0$ process. Thus, I conclude that the dark state of the divacancy is $V_{Si}V_C^-$ [P1].

(b) **Quenching mechanism at elevated temperatures ($T > 0$ K).**

At elevated temperatures the reionization process of $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ may be influenced by phonons lowering the excitation threshold energies. I calculated temperature dependent HR spectra of the $V_{Si}V_C^- \rightarrow V_{Si}V_C^0$ for all $V_{Si}V_C$ defect configurations by means of density functional plane wave supercell calculations. According to my results, reionization with the cumulative transition probability of half of the DW factor can be achieved at room temperature (300 K) by employing the excitation energy used for the optical readout $V_{Si}V_C^0$ qubit. This enables the charge state control and simultaneous qubit manipulation of $V_{Si}V_C^0$.

2. Nitrogen-vacancy (NV) center in SiC.

Negatively charged nitrogen-vacancy defect (NV center) in 4H SiC is promising solid state qubit candidate. NV center in 4H SiC forms 4 different defect configurations as: *hh*, *kk*, *hk*, *kh* configurations, where the first letter denotes the site of N atom (N_C) while the second stands for the site of Si vacancy (V_{Si}). Novel PL and EPR centers have been assigned to NV centers in 4H SiC and all four signals have been observed [von Bardeleben et al., 2015, von Bardeleben et al., 2016, Zargaleh et al., 2016]. The measured ZPLs are 0.998 eV (PLX1), 0.999 eV (PLX2), 1.014 eV (PLX3) and 1.051 eV (PLX4) while the corresponding D constant values are 1193 MHz, 1270 MHz, 1313 MHz and 1328 MHz.

Magneto-optical properties provide unambiguous fingerprints for defect indentification. Hence I determined the ZPLs, constants of hyperfine coupling assuming ^{29}Si and ^{13}C isotopes ($A_\parallel$, $A_\perp$), and zero-field splitting (ZFS) parameters D and E by means of density functional plane wave calculations.

I also investigated the fabrication procedure of NV centers in 4H SiC. To this end I considered two mature processes: (i) the chemical vapor deposition (CVD) and the (ii) ion implantation procedure followed by annealing at high temperatures. While the (i) procedure was approximated as an equilibrium process, the (ii) one was treated as non-equilibrium mechanism in my calculations. Regarding case (i), I considered maximum N concentration. In this way intrinsic defects, substitutional N defects (N_C), N_CV_{Si} defects

and N-precipitate [$(N_C)_4V_{Si}$] may be formed. For calculating the concentrations I considered the usually applied temperatures (between 1600 °C and 2000 °C) and applied the charge neutrality equation with assuming thermal equilibrium of the considered defects in 4H SiC. Via applying the (ii) procedure, vacancies and interstitial atoms are possibly created in the lattice of 4H SiC. Follow-up annealing mobilizes these defects giving rise to different defect complexes. According to experiments divacancy in its neutral charge state is always formed besides NV center as a result of process (ii) [von Bardeleben et al., 2015, von Bardeleben et al., 2016]. In order to predict concentration relations, I calculated the binding energies of these two complexes.

In quantum technological aspects, optical stability (or photostability) of NV center is crucial for accurate initialization and read-out processes realized via optical excitation. This can be violated by two phenomena that may emerge under illumination: by ionization of NV center, or by the Stark effect causing spectral diffusion, i.e. shift of the corresponding ZPLs. Stark effect is given rise by charge fluctuations originating from ionization of other defects around the NV center. The most important defect accompanying NV center in ion-implanted 4H SiC samples is the divacancy. In this context I investigated two-photon processes which possibly occurs in confocal microscopy measurements and may result in ionization of the NV center and/or the divacancy.

For the calculations I employed PBE and HSE06 functionals and 576 atom 4H SiC supercell. I group my results into 3 thesis points.

(a) **Identification of defect configurations in the 4H polytype.**

I unambiguously identified the NV center configurations via the comparison of the calculated and observed ZPLs and D constants. The calculated ZPL values are 0.966 eV (hh), 1.018 eV (kk), 1.039 eV (hk) and 1.056 eV (kh) while those for the D constants are 1427 MHz (hh), 1377 MHz (kk), 1331 MHz (hk) and 1404 MHz (kh). Based on the obtained numerical results for the magneto-optical parameters I associated the corresponding PL signals (PLX1-4) [Zargaleh et al., 2016] as hh - PLX1; kk - PLX2; hk - PLX3; kh - PLX4 [P2, P3].

(b) **Formation of NV center in 4H SiC.**

My results for the simulation of CVD preparation imply that concentrations of intrinsic defects are smaller by orders of magnitude than those of the other investigated defects. Concentration of the neutral $(N_C)_4V_{Si}$ ($S = 0$ spin state) and N_C ($S = \frac{1}{2}$ spin state) is higher by about 9 and by about 7 orders of magnitude, respectively, than that of NV center. While the former defect is spinless and hence indifferent, dense spin bath is established by

the neutral N_C in such high concentration being detrimental for the spin manipulation of NV center. Concentration of the doubly negative $N_C V_{Si}$ ($S = \frac{1}{2}$ spin state) defect is also slightly higher than that of NV center introducing a further drawback with respect to the potential applications. Based on the results for implantation, binding energy of divacancy is higher than that of NV center by about $\approx$ 2-3 eV depending on the position of the Fermi level in 4H SiC. As a consequence, divacancy always forms besides NV center in samples prepared with implantation or irradiation.

Consequently, I conclude that preparation with ion implantation is the better strategy for fabrication NV center. In particular highly N-doped 4H SiC sample prepared by ion implantation may result predominant accumulation of NV center over divacancy [P3].

(c) **Photostability of NV center in ion-implanted 4H SiC.**

According to my results based on HSE06 hybrid density functional theory calculations, ionization threshold of NV center i.e. energy of the NV(-) $\rightarrow$ NV(0) transition is around $\approx$ 1.7-1.8 eV in 4H SiC. On the other hand ionization threshold of neutral divacancy, i.e. energy of the $V_{Si}V_C^0 \rightarrow V_{Si}V_C^-$ process is about $\approx$ 2.1 eV. Excitation energy of NV center is $\approx$ 1.0 eV while that of divacancy is $\approx$ 1.1 eV. Consequently, by photo-excitation into the phonon-sideband (PSB) of NV center ($\geq$ 1.1 eV) two-photon absoprtion process may occur for both NV center and divacancy resulting in ionization of both defects, which in turn reduces the photostability of the NV center.

Based on my results I conclude that the best strategy to maintain the photostability of NV center is to employ excitation energies below 1.1 eV avoiding the divacancy to be ionized by two-photon processes [P3].

3. Vanadium point defect in 4H SiC

Vanadium impurity in 4H SiC has been proven to be a suitable single photon emitter candidate for quantum communication owing to its NIR emission falling into the O-band (1260-1360 nm) of telecom wavelength region [Spindlberger et al., 2019, Wolfowicz et al., 2019]. Preliminary studies reveal that vanadium forms substitutional defects at a Si site [Ivády et al., 2011] that are neutral in the investigated intrinsic 4H samples (Van_{Si}^0). Structure of the 4H lattice support formation of two kinds of Van_{Si} defect as $Van_{Si}(h/k)$ depending on the occupied hexagonal/cubic site. Based on previous experimental results electronic structure of Van_{Si}^0 defects in 6H SiC has already been described by employing crystal field theory [Kaufmann et al., 1997, Kunzer et al., 1993] that can be expected to closely related to that of Van_{Si}^0 defects in 4H SiC. Novel PL spectra [Spindlberger et al., 2019] of Van_{Si}^0

in 4H SiC are also available, where two ZPLs have been observed: the so-called α line at 0.971 eV and the β line at 0.929 eV. The α line shows single exponential decay, however the β line exhibits double exponential decay. Moreover, several sharp lines take place in the PSB.

On the other hand, results from *ab initio* calculations were not available before neither for 6H nor for 4H SiC. Therefore, I calculated the electronic structure of $\mathrm{Van}^0_{\mathrm{Si}}$ defects in 4H SiC by means of the HSE06+V_w technique [Ivády et al., 2014] assisted with PBE [Perdew et al., 1996] functional in a 576 atom supercell.

I provide my results in the following 4 thesis points.

(a) **Electronic structure of vanadium defect in 4H SiC**

Vanadium related ingap states are formed by the atomic d-orbitals which remain strongly localized even in the 4H SiC lattice. Characters of the one-electron states in energy order are $e(1)e(0)a_1(0)$ for $\mathrm{Van}^0_{\mathrm{Si}}(k)$ and $e(1)a_1(0)e(0)$ for $\mathrm{Van}^0_{\mathrm{Si}}(h)$, where the occupation numbers are denoted in brackets. Consequently, $\mathrm{Van}^0_{\mathrm{Si}}$ introduces ^2E spin doublet ground state. The $a_1(0)$ and the higher-energy $e(0)$ states are resonant with the conduction band edge in both cases, however for $\mathrm{Van}^0_{\mathrm{Si}}(k)$ the a_1 state shifts upward due to the stronger crystal field confinement along the c-axis [P4].

(b) **Identification of the vanadium-related ZPLs.**

Based on the differences between the $\mathrm{Van}^0_{\mathrm{Si}}(h)$- and $\mathrm{Van}^0_{\mathrm{Si}}(k)$-related electronic structures and the calculated ZPLs I identified the $\mathrm{Van}^0_{\mathrm{Si}}$-related ZPLs. Accordingly, calculated ZPL energies are 0.785 eV for $\mathrm{Van}^0_{\mathrm{Si}}(h)$ and 0.845 eV for $\mathrm{Van}^0_{\mathrm{Si}}(k)$. Consequently, I assign the lower-in-energy β line to $\mathrm{Van}^0_{\mathrm{Si}}(h)$ and the α line to $\mathrm{Van}^0_{\mathrm{Si}}(k)$ which contradicts the previous interpretations in the literature [Kaufmann et al., 1997, Kunzer et al., 1993] [P4].

(c) **Identification of the vanadium-related features in the PL spectrum.**

I calculated the energy and localization of the $\mathrm{Van}_{\mathrm{Si}}$-related local phonon modes in the PSB giving the possibility to identify the other features present in the PL spectrum at energies of 0.840 eV - β (4 × C); 0.860 eV - β [4 × C + $\mathrm{Van}_{\mathrm{Si}}(h)$]; 0.872 eV - α [4 × C + $\mathrm{Van}_{\mathrm{Si}}(k)$)]; 0.882 eV - α (4 × C); 0.902 eV - β [$\mathrm{Van}_{\mathrm{Si}}(h)$]; 0.949 eV - α [$\mathrm{Van}_{\mathrm{Si}}(k)$]. Atoms vibrating in the corresponding phonon modes are denoted in brackets as $\mathrm{Van}_{\mathrm{Si}}$ - Vanadium atom; 4 × C - neighboring 4 C atom [P4].

(d) **Radiative lifetimes**

By DFT calculations I verified that the biexponential decay of the β singlet line is giving rise by the α-related PSB extending beyond the β ZPL. Decay of the ZPL lines are governed by the corresponding total fluorescence decay lifetimes which had been determined from

experiments. In addition, I calculated the radiative lifetimes of both Van_{Si}^{0} defects that is 704 ns for $Van_{Si}(k)$ (α) and 277 ns for $Van_{Si}(h)$ (β). By taking the observed optical lifetimes and computed radiative lifetimes, the non-radiative lifetimes can also be determined that are 212 ns for $Van_{Si}(k)$ (α) and 47 ns for $Van_{Si}(h)$ (β) [P4].

Impact

Results achieved during my PhD course may contribute to the fabrication and application of paramagnetic point defects embedded in SiC as quantum bits and single photon sources.

Publications related to the thesis points

[P1] B. Magnusson, N. T. Son, A. Csóré, A. Gällström, T. Ohshima, A. Gali, I. G. Ivanov, Excitation properties of the divacancy in 4H-SiC, Phys. Rev. B 98, 195202 (2018)

[P2] H. J. von Bardeleben, J. L. Cantin, A. Csóré, A. Gali, E. Rauls, and U. Gerstmann, NV centers in 3C, 4H, and 6H silicon carbide: A variable platform for solid-state qubits and nanosensors, Phys. Rev. B 94, 121202(R), (2016)

[P3] A. Csóré, H. J. von Bardeleben, J. L. Cantin, A. Gali, Characterization and formation of NV centers in 3C, 4H and 6H SiC: an ab initio study, Phys. Rev. B, 96, 085204 (2017)

[P4] L. Spindlberger, A. Csóré, G. Thiering, S. Putz, R. Karhu, J. Ul Hassan, N. T. Son, T. Fromherz, A. Gali and M. Trupke, Optical properties of vanadium in 4H silicon carbide for quantum technology, Phys. Rev. Applied 12, 014015 (2019)

Further publicatons

[P5] A. Csóré, A. Gällström, E. Janzén, A. Gali, Investigation of Mo Defects in 4H-SiC by Means of Density Functional Theory, Mater. Sci. Forum 858, pp. 261-264 (2016)

[P6] A. Csóré, A. Gali, Density functional theory on NV center in 4H SiC, Mater. Sci. Forum 897, pp. 269-274 (2017)

[P7] A. Csóré, B. Magnusson, N. T. Son, A. Gällström, T. Ohshima, I. G. Ivanov, A. Gali, First-principles study on photoluminescence quenching of divacancy in 4H SiC, Mater. Sci. Forum 963, pp. 714-717 (2019)

[P8] A. Csóré, A. Gali, *Ab initio* determination of pseudospin for paramagnetic defects in SiC, arXiv:1909.11587 [quant-ph]

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