Variability in Si/SiGe and Si/SiO_2 Spin Qubits due to Interfacial Disorder

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***Abstract*—Silicon provides a promising platform to host solid-state spin qubits owing to long coherence times through isotopic purification and the highly advanced level of development in material processing and fabrication techniques. In practice, the uniformity of spin qubits is limited by uncontrolled atomistic fluctuations at the interface between the semiconductor and the confining material. Maintaining a qubit fidelity well above the quantum fault-tolerance threshold is only possible for a clean and well-defined two state system and accounting for the specific parameters of each qubit would quickly become a bottleneck for any large-scale quantum computer. Despite extensive efforts in the modeling of qubits, a detailed understanding of the impact of interfacial disorder on the atomic scale is still missing. Within this work, we model realistic Si-based 3D interface structures by *ab initio* calculations. We improve current modeling approaches by considering a disordered atomistic environment and extract important qubit parameters like valley and spin splittings from these models.**

I. Introduction

Since the theoretical foundation for reliable quantum computation has been laid down decades ago, the race for finding a robust and scalable realization of qubits, the basic storage devices for quantum information, is currently ongoing around the world. Realizations in superconductors, cold atoms, and trapped ions have been demonstrated, however, these require immense efforts and are not easily miniaturizable [1]. Solid-state spin qubits on the other hand, including their most popular representative in silicon (Si), use material systems which have been utilized in microelectronic devices for decades, and can thus build upon the vast experience available in semiconductor manufacturing. This allows to employ established fabrication techniques with well-proven processes and materials. Furthermore, Si promises long spin coherence times due to its nuclear-spin-free isotope ^{28}Si, and can be further enriched to minimize hyperfine interactions with the carrier spin [2], [3]. Building on MOS fabrication techniques, Si/SiO_2 qubits have already been realized [4], [5]. However, these qubits suffer from short coherence times even for purified ^{28}Si due to charge noise and interactions with the oxygen nuclei at the interface. Engineering the environment in order to obtain more stable qubit states has recently led to the use of SiGe as the confining material, effectively prolonging decoherence times [6]. Also, universal control of a six-qubit quantum processor in Si/SiGe has been achieved [7].

Previous theoretical approaches did not explicitely consider atomistic disorder but rather assumed crystalline interfaces [8]–[10]. Mesoscopic device simulations have been done by means of tight-binding (TB) and $k \cdot p$ methods but were similarly limited to overidealized interfaces [11] or, at most, included interface roughness as one source of disorder [12]. Compared to these methods, density functional theory (DFT) is limited to smaller simulation cells. It is, however, well suited to understand the impact of local disorder at the interface directly from first principles without any (semi)empirical parameters.

In this work, we report variabilities in valley and spin splittings of Si based qubits in which the spin/charge carrier is confined by SiO_2 and SiGe, respectively. We employ DFT calculations and study realistically disordered, yet qualitatively similar interface structures. We show that the splittings are distributed by about one order of magnitude, emphasizing the relevance of atomistic details for reliable quantum devices and the potential of theoretical investigations for future developments. Given this large spread, accurate modeling, similar to technology computer-aided design (TCAD) in conventional semiconductor devices, will be indispensable for the design and exploration of scalable qubits.

II. Models and Materials

We model the two most important types of Si based qubits in which the conduction band valley electron is confined in a Si layer (thickness 2–14 nm) by SiGe or SiO_2, respectively. We focus on the effect of disordered interfaces, that is, the structures do not show any other sources of variability such as long-range interface roughness, steps or atomic defects. Due to periodic boundary conditions, the actual model forms a 1D well, as shown in Fig. 2a. The interfaces were mirrored about an inversion center as can be seen in the cross-section averaged Hartree potential (that is the classical potential of ions and valence electrons combined plus possible contributions from external electric fields) of the simulation cell.

The Si/SiGe structure has a pseudomorphic interface. When growing Si on SiGe, the Si layer adapts to the SiGe substrate and is thus strained due to a lattice mismatch of up to 4% (depending on the Ge concentration). The strain assists the emergence of a clean qubit in two ways: first, it lifts the degeneracy of four out of the six conduction band valleys.

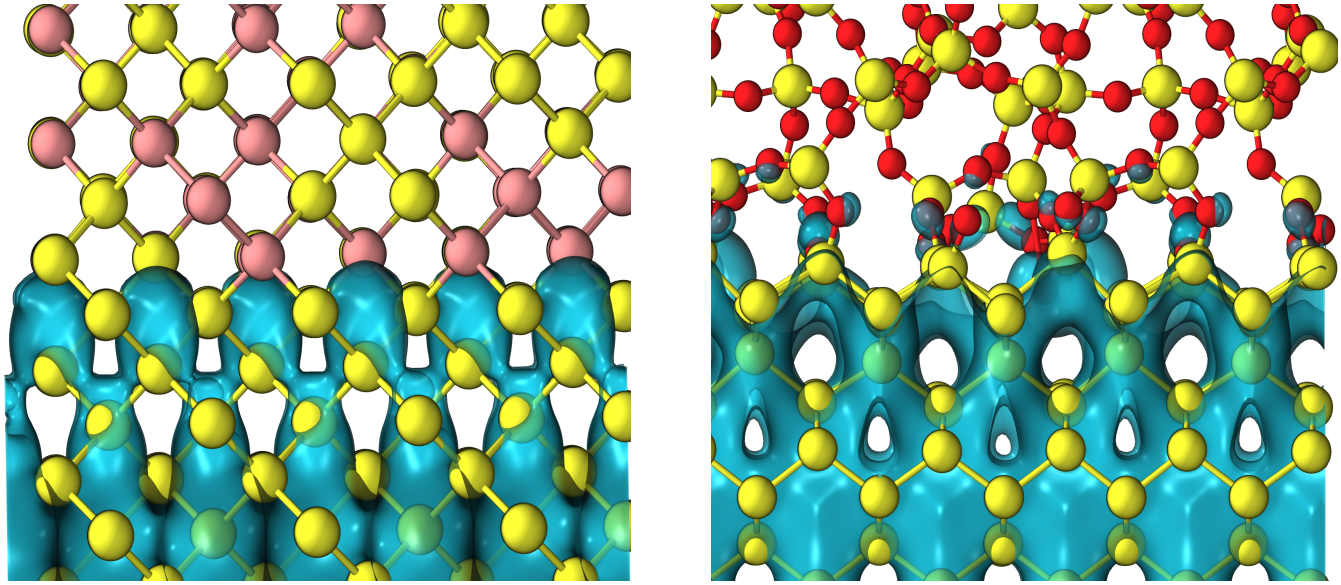

Fig. 1. Representative interfaces between Si and SiGe (left) and amorphous SiO_2 (right). The electron valley wavefunction from DFT is depicted as a blue isosurface. Si, Ge and O atoms are depicted in yellow, pink and red, respectively.

Second, since the band alignment between Si and SiGe is strain-dependent, strained Si between relaxed SiGe leads to a quantum well in the Si layer. We assume a Ge concentration of 30% in the SiGe layer. Disorder is introduced as random distributions of Ge atoms in the SiGe layer resulting in local fluctuations of the Ge concentration in the $Si_{0.7}Ge_{0.3}$ part.

The Si/SiO_2 interface is amorphous and as such yields a considerable inherent disorder and surface roughness. The realistic interface structures were generated by *ab-initio* molecular dynamics simulations and show a 0.5 nm thick transition layer between Si and SiO_2 [13]. The oxide layer is 1.5 nm thick and passivated with H on both sides of the supercell. A vacuum of 2 nm is introduced in order to construct a slab model of the $SiO_2/Si/SiO_2$ heterostructure.

The electronic structure of these interface models was obtained by using DFT as implemented in CP2K [14]. The orbitals are expanded in the GTH basis set [15] and the exchange-correlation energy was considered within the semilocal GGA-PBE approximation [16].

III. Valley Splitting

Electrons in pristine Si occupy 6-fold degenerate valleys in the conduction band. When applying an in-plane strain to a Si layer, this degeneracy is partially lifted, resulting in a twofold degenerate ground state. The valley states in z-confined Si can be described as [17]

$$\Psi_j(z) = \mathrm{e}^{ik_j z} u_{\mathbf{k}_j} F(z) \tag{1}$$

with the valley wave vector $k_{j=\pm} = \pm\, 0.85\, \frac{2\pi}{a}$ ($a = 0.54$ nm is the lattice constant of Si), the bloch function $\mathrm{e}^{ik_j z} u_{\mathbf{k}_j}$ and the well envelope $F(z)$. The quantum well states then take the form

$$\Psi_\pm = \frac{1}{\sqrt{2}} \left(\Psi_{+z}(z) \pm \mathrm{e}^{i\theta} \Psi_{-z}(z) \right) \tag{2}$$

where θ denotes a phase shift between the valley states. An energy splitting between the two states in (2) is obtained by the sharp spatial confinement in z direction – e.g. by interfaces to other materials [8]. The resulting two energy levels can be used as a *valley qubit*. The valley splitting depends on geometric parameters like the thickness of the semiconductor layer [8]. However, also the importance of disorder and atomic fluctuations at the interface have recently gained attention in the spin qubit community [18], [19].

Here, we compare the valley splittings obtained by DFT to a TB approach from [8]. The results are summarized in Fig. 2. The simple 1D two-band second-nearest neighbor Si TB model considers hard wall boundary conditions ($|\Psi_\pm(z)|^2$ vanishes at the interface – infinite well) or soft wall boundary conditions (the wavefunction extends in confining layers whose on-site energies are increased by 3.2 eV, which is the experimental band offset of SiO_2.) This model qualitatively captures the trends for the well width dependence as well as the influence of the electric field. In contrast to our DFT models, the basic TB approach does not take the disorder of the interface into account. Thus, there is no spread in the TB results. As can be seen in Fig. 2b, the VS is decreases substantially for certain numbers of Si layers in the well. This results from an interference between the two interfaces: without an electric field, the valley states couple with both interfaces [8]. In order to probe single interfaces, we push the qubit wavefuntion to one interface by applying an electric field in z direction. Thus, for a given field, there is a Si width beyond which the VS stays constant (blue line in Fig. 2b). Extending the Si width does not change the wavefunction any more. The interference effect can also be observed in the electric field dependence (Fig. 2c) which results in a power-law increase above a certain value of the electric field, indicating the single-interface regime. As can be seen in Fig 2b and 2c, the atomistic disorder yields a wide distribution of valley splittings by about one order of magnitude, in agreement with a wide range of experimentally observed valley splittings in Si/SiO_2 and Si/SiGe [20]–[22].

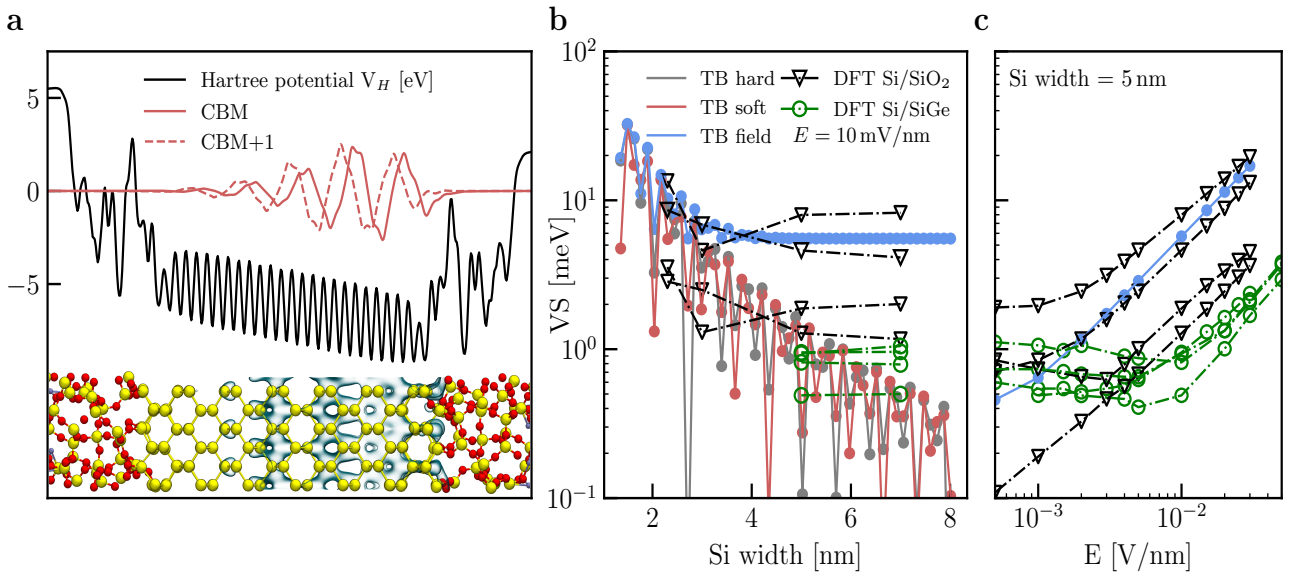

Fig. 2. **a** The two valley wavefunctions averaged over the cross section confined in a 2D quantum well represented by the Hartree potential. The z and $-z$ valley states are phase shifted in real space, which leads to different penetration depths into the confining barrier and introduces the valley splitting. The well is tilted due to the application of an electric field. **b** The valley splitting depends on the width of the Si layer. The electric field can push the wavefunction against one interface, so that the splitting stays constant even if the well width is further increased. The dot-dashed lines are DFT calculated values at $E = 10\,\mathrm{mV/nm}$ for qualitatively similar interfaces which are distributed over one order of magnitude. **c** Electric field dependence of the valley splitting for a well width of 5 nm. The onset of the single interface regime is indicated by a power-law increase (note the log-log scale). For comparison with experimental values of qubit devices, one needs to consider the strongly influential Si width.

IV. Spin splitting

So far, we did not consider spin in the simple band description above, thus each band would be spin degenerate. This degeneracy is lifted by spin-orbit coupling (SOC). Solely based on symmetry arguments, it can be shown that only two types of spin Hamiltonians are allowed which lead to Rashba [23] and Dresselhaus [24] spin-orbit couplings, respectively:

$$H_\mathrm{R} = \alpha_\mathrm{R}(\sigma_x k_y - \sigma_y k_x) \tag{3}$$

$$H_\mathrm{D} = \alpha_\mathrm{D}(\sigma_x k_x - \sigma_y k_y) \tag{4}$$

with the Pauli matrices σ_i. The Dresselhaus-like term usually arises due to inversion asymmetry in a crystal unit cell (and should thus be absent in Si). However, in this configuration the Dresselhaus term stems from breaking the inversion symmetry of the supercell by a finite electric field of 10mV/nm [10].

In a *spin qubit*, the quantum state is then encoded in two spin states of an electron in a magnetic field. In this case, a large valley splitting is essential, otherwise the remaining valley state would act as a leakage channel for the quantum state information. Furthermore, knowledge about the SOC in a spin qubit could be essential for driving qubit state transitions by oscillating electric fields, a technique termed electron dipole spin resonance (EDSR) [6]. Compared to holes (in hole spin qubits), the SOC is rather weak in the conduction band, which limits the electrical manipulation of electron spin qubits. For this reason, an enhancement of the SOC is highly desirable, and can be achieved either by placing micro-magnets in the vicinity of the qubit [25] or by engineering the shape of the quantum wells [26]. However, our results imply that SOC in electron spin qubit devices could also be enhanced by modifying the microscopic structure of the interface.

As shown in Fig. 3, we find a linear-in-k spin splitting for small k. For larger k, the splitting can be fitted to a cubic term as expected by the theory. This is depicted in the inset. The spin splitting was calculated for k points along the [110] and [1$\bar{1}$0] directions, respectively. This allows for simple extraction of the Rashba and Dresselhaus coefficients α_R and α_D by symmetry arguments. With respect to the rotated k-axis, the Dresselhaus term gives the symmetric contributions while the Rashba term results in anti-symmetric contributions. The extracted values for α_R vary between 0.4 and 0.8 µeVnm and for α_D from 37 to 664 µeVnm. The dominating Dresselhaus contributions are in rough agreement with earlier TB results from [9], but our values vary by more than one order of magnitude purely due to the interfacial disorder.

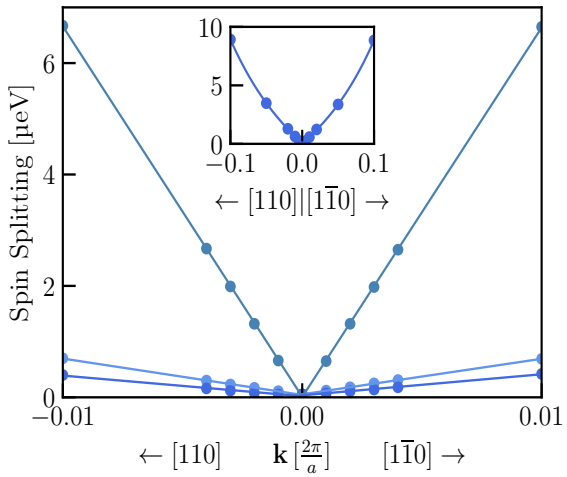

Fig. 3. Spin splitting of the lowest valley state for $\mathbf{k} = [k_x, k_y]$ along the in-plane directions [110] and [1$\bar{1}$0] at $E = 10\,\mathrm{mV/nm}$ for Si/SiO$_2$. The splitting is linear in k for small k and cubic for larger k as shown in the inset.

V. Conclusions

In this work, we established an *ab-initio* based modeling approach to extract important qubit parameters from atomistic heterostructures. This approach can be applied to various material systems as demonstrated for the currently most popular types of Si based semiconductor qubits, Si/SiO_2 and Si/SiGe. Our first principles based approach combined with realistic, atomistic interface structures goes beyond modeling techniques that have been used so far, most importantly TB and $k \cdot p$, and allows to investigate the impact of atomistic disorder in considerable detail. Stress and strain, disorder at the interface, and atomistic fluctuations are inherently captured by our DFT model.

By means of this approach, we find wide distributions of valley and spin splittings by more than one order of magnitude, which can be attributed to the variability of the interface due to intrinsic disorder of the confining materials. Ultimately, the valley and spin splittings determine the qubit state manipulation process [12]. In a reliable quantum processor, thousands of individually controllable qubits need to be integrated on one chip. Adressing the variability of each of the qubits would be extremely challenging in practice. Thus, our calculations underline the importance of reproducible interfaces for scalable quantum devices. On the other hand, our results imply that valley- and spin splittings could be tuned by engineering the interfaces to the confining materials.

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References

[1] G. Popkin, "Quest for qubits," *Science*, vol. 354, no. 6316, pp. 1090–1093, 2016. [Online]. Available: https://www.science.org/doi/abs/10.1126/science.354.6316.1090

[2] F. A. Zwanenburg, A. S. Dzurak *et al.*, "Silicon quantum electronics," *Rev. Mod. Phys.*, vol. 85, pp. 961–1019, 2013.

[3] G. Burkard, T. D. Ladd, J. M. Nichol, A. Pan, and J. R. Petta, "Semiconductor spin qubits," 2021. [Online]. Available: https://arxiv.org/abs/2112.08863

[4] R. Maurand *et al.*, "A CMOS Si spin qubit," *Nature Communications*, vol. 7, p. 13575, 2016.

[5] B. Voisin, V.-H. Nguyen *et al.*, "Few-electron edge-state quantum dots in a silicon nanowire field-effect transistor," *Nano Letters*, vol. 14, no. 4, pp. 2094–2098, 2014. [Online]. Available: https://doi.org/10.1021/nl500299h

[6] E. Kawakami, P. Scarlino *et al.*, "Electrical control of a long-lived spin qubit in a Si/SiGe quantum dot," *Nature Nanotechnology*, vol. 9, no. 9, pp. 666–670, 2014. [Online]. Available: https://doi.org/10.1038/nnano.2014.153

[7] S. G. J. Philips, M. T. Madzik *et al.*, "Universal control of a six-qubit quantum processor in silicon," *Nature*, vol. 609, no. 7929, pp. 919–924, 2022. [Online]. Available: https://doi.org/10.1038/s41586-022-05117-x

[8] T. B. Boykin, G. Klimeck, M. Friesen, S. N. Coppersmith, P. von Allmen, F. Oyafuso, and S. Lee, "Valley splitting in low-density quantum-confined heterostructures studied using tight-binding models," *Phys. Rev. B*, vol. 70, p. 165325, 2004. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.70.165325

[9] M. Prada, G. Klimeck, and R. Joynt, "Spin–orbit splittings in Si/SiGe quantum wells: from ideal Si membranes to realistic heterostructures," *New Journal of Physics*, vol. 13, no. 1, p. 013009, 2011. [Online]. Available: https://dx.doi.org/10.1088/1367-2630/13/1/013009

[10] M. O. Nestoklon, E. L. Ivchenko, J.-M. Jancu, and P. Voisin, "Electric field effect on electron spin splitting in SiGe/Si quantum wells," *Phys. Rev. B*, vol. 77, p. 155328, 2008. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.77.155328

[11] L. Bourdet and Y.-M. Niquet, "All-electrical manipulation of silicon spin qubits with tunable spin-valley mixing," *Phys. Rev. B*, vol. 97, p. 155433, 2018. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.97.155433

[12] B. Martinez and Y.-M. Niquet, "Variability of electron and hole spin qubits due to interface roughness and charge traps," *Phys. Rev. Appl.*, vol. 17, p. 024022, 2022. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevApplied.17.024022

[13] L. Cvitkovich, D. Waldhör, A.-M. El-Sayed, M. Jech, C. Wilhelmer, and T. Grasser, "Dynamic modeling of Si(100) thermal oxidation: Oxidation mechanisms and realistic amorphous interface generation," *Applied Surface Science*, vol. 610, p. 155378, 2023. [Online]. Available: https://www.sciencedirect.com/science/article/pii/S0169433222029063

[14] T. Kühne, M. Iannuzzi *et al.*, "CP2K: An electronic structure and molecular dynamics software package - quickstep: Efficient and accurate electronic structure calculations," *J. Chem. Phys.*, vol. 152, pp. 580–586, 2020.

[15] S. Goedecker, M. Teter, and J. Hutter, "Separable dual-space gaussian pseudopotentials," *Phys. Rev. B*, vol. 54, pp. 1703–1710, 1996. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.54.1703

[16] J. P. Perdew, K. Burke, and M. Ernzerhof, "Generalized gradient approximation made simple," *Phys. Rev. Lett.*, vol. 77, pp. 3865–3868, 1996. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevLett.77.3865

[17] M. Friesen, S. Chutia, C. Tahan, and S. N. Coppersmith, "Valley splitting theory of SiGe/Si/SiGe quantum wells," *Phys. Rev. B*, vol. 75, p. 115318, 2007. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.75.115318

[18] J. P. Dodson, H. E. Ercan *et al.*, "How valley-orbit states in silicon quantum dots probe quantum well interfaces," *Phys. Rev. Lett.*, vol. 128, p. 146802, 2022. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevLett.128.146802

[19] B. Wuetz *et al.*, "Atomic fluctuations lifting the energy degeneracy in Si/SiGe quantum dots," *Nature Communications*, vol. 13, no. 1, p. 7730, 2022. [Online]. Available: https://doi.org/10.1038/s41467-022-35458-0

[20] X. Mi, C. G. Péterfalvi, G. Burkard, and J. R. Petta, "High-resolution valley spectroscopy of Si quantum dots," *Phys. Rev. Lett.*, vol. 119, p. 176803, 2017. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevLett.119.176803

[21] Z. Shi, C. B. Simmons *et al.*, "Tunable singlet-triplet splitting in a few-electron Si/SiGe quantum dot," *Applied Physics Letters*, vol. 99, no. 23, 2011, 233108. [Online]. Available: https://doi.org/10.1063/1.3666232

[22] C. B. Simmons, T. S. Koh *et al.*, "Pauli spin blockade and lifetime-enhanced transport in a Si/SiGe double quantum dot," *Phys. Rev. B*, vol. 82, p. 245312, 2010. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.82.245312

[23] Y. A. Bychkov and E. I. Rashba, "Oscillatory effects and the magnetic susceptibility of carriers in inversion layers," *Journal of Physics C: Solid State Physics*, vol. 17, no. 33, p. 6039, 1984. [Online]. Available: https://dx.doi.org/10.1088/0022-3719/17/33/015

[24] G. Dresselhaus, "Spin-orbit coupling effects in zinc blende structures," *Phys. Rev.*, vol. 100, pp. 580–586, 1955. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRev.100.580

[25] M. Veldhorst, J. C. C. Hwang *et al.*, "An addressable quantum dot qubit with fault-tolerant control-fidelity," *Nature Nanotechnology*, vol. 9, no. 12, pp. 981–985, 2014. [Online]. Available: https://doi.org/10.1038/nnano.2014.216

[26] B. D. Woods, M. A. Eriksson, R. Joynt, and M. Friesen, "Spin-orbit enhancement in Si/SiGe heterostructures with oscillating Ge concentration," *Phys. Rev. B*, vol. 107, p. 035418, 2023. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.107.035418