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Master's Thesis

Acoustic Manipulation of Spin Qudits on 6H-SiC

submitted by

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prepared in cooperation with

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Berlin, August 24, 2026

Statement of Authorship

I hereby declare that I have written this master's thesis independently and have used no sources or aids other than those stated. All passages taken from published and unpublished sources, whether quoted directly or paraphrased, have been clearly indicated as such.

Berlin, August 24, 2026

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List of Abbreviations

Abbreviation	Meaning
3C-SiC	Cubic 3C polytype of silicon carbide
4H-SiC	Hexagonal 4H polytype of silicon carbide
6H-SiC	Hexagonal 6H polytype of silicon carbide
AlN	Aluminium nitride
BAW	Bulk acoustic wave
CMOS	Complementary metal–oxide–semiconductor
DC	Direct current
ES	Excited state
ESLAC	Excited-state level anticrossing
FWHM	Full width at half maximum
GaN	Gallium nitride
GS	Ground state
HDF5	Hierarchical Data Format version 5
HZDR	Helmholtz-Zentrum Dresden-Rossendorf
IDT	Interdigital transducer
MEMS	Micro-electro-mechanical systems
MIF	Metal-ion-free
MoS ₂	Molybdenum disulfide
MW	Microwave
ODMR	Optically detected magnetic resonance
PL	Photoluminescence
RF	Radio frequency
RMS	Root mean square

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Abbreviation	Meaning
RPM	Revolutions per minute
SAW	Surface acoustic wave
SF	Single finger
SiC	Silicon carbide
SiO ₂	Silicon dioxide
VNA	Vector network analyser
XRD	X-ray diffraction
ZnO	Zinc oxide

Chapter 1

Introduction

Over the recent years, the research and interest on Silicon carbide (SiC) and its polymorphs have been increasing due to its remarkable electric and optical tuneability and radiation stability[4]. For quantum technologies, one of the most interesting point of interest is the negatively charged silicon vacancy V_{Si} which results from a missing silicon atom in the crystal lattice[4][5]. The most common SiC crystal poly-types are 3C-SiC, 4H-SiC, and 6H-SiC, where “C” represents a cubic crystal structure and “H” represents a hexagonal crystal structure[6]. The colour centres on these poly-types are a good single photon source and also particularly interesting due to their long spin coherence time as well as an effective spin qubit even at room temperature[7].

Since the last two decades SiC structures have gained vast interests and popularity with their colour centres which can be beneficial in quantum technologies[8][9].

In the previous literature the structure of 4H-SiC[5][10][11], 6H-SiC[10][5][1][12] have been studied; spin-acoustic coupling on 4H-SiC[13][14] have been demonstrated.

This thesis reviews the literature on SiC polytypes, surface acoustic waves (SAWs), and the acoustic manipulation of colour centres. The first part acts as a literature review which introduces the structural properties of different SiC polytypes, the operation of interdigital transducers (IDTs), manipulation of the colour centres with these IDTs via phonon-photon coupling and the current state of the art. The second part presents the experimental work performed on the acoustic manipulation of spin qubits in 6H-SiC, including the laboratory procedure, the methods used, and the results obtained from the measurements.

1.1 Research Gap and Motivation of This Thesis

Optically active point defects in wide-bandgap semiconductors have emerged as promising solid-state platforms for quantum technologies. Far from being merely detrimental imperfections, selected defect centres can provide well-defined electronic spin states, stable optical transitions, and long spin coherence times. These properties make them suitable for applications such as single-photon generation, quantum sensing, spin-based quantum memories, and spin-photon interfaces. A particularly important class of such systems is formed by optically addressable spin centres, whose spin states can be initialized and read out using light. Their spin dynamics are commonly investigated through optically detected magnetic resonance (ODMR), where changes in the photoluminescence intensity provide information about magnetic-resonance-driven transitions between spin sublevels [4].

Silicon carbide (SiC) is especially attractive in this context because it combines mature semiconductor processing compatibility with the presence of highly coherent and optically active defect centres. As a wide-bandgap semiconductor compatible with established microfabrication and CMOS-related technologies, SiC offers a promising route toward scalable and integrated quantum devices. Among the relevant defects in SiC, the negatively charged silicon vacancy, V_{Si}^- , is of particular interest. Its spin-3/2 ground state provides a four-level spin system, or spin qudit, which offers richer functionality than a simple two-level qubit and can be useful for robust quantum information protocols [1].

1.1.1 Motivation for acoustic manipulation in 6H-SiC

Conventional spin manipulation is typically performed using microwave magnetic fields. However, acoustic control based on surface acoustic waves (SAWs) offers an alternative and highly attractive approach. SAWs generate dynamic strain fields that propagate along the surface of a material and can interact with quantum systems located within their propagation path. Through spin-phonon coupling, these dynamic strain fields can drive transitions between spin states, enabling coherent spin manipulation using sound rather than purely electromagnetic fields.

Previous work on V_{Si}^- centres in 4H-SiC [14][13][15] and divacancy $V_{\text{Si}}^- V_{\text{C}}^-$ centres [16] has demonstrated that SAWs can provide an efficient mechanism for controlling spin dynamics. This establishes acoustic spin control as a powerful tool for coupling mechanical excitations to solid-state spin systems. Nevertheless, the extension of this approach to other SiC polytypes and defect configurations remains comparatively unexplored. In particular, 6H-SiC hosts distinct silicon-vacancy-related centres, including the V3 centre, whose optical and spin properties make it a promising candidate for advanced quantum-photonics applications.

The V3 centre in 6H-SiC is especially interesting because of its potential relevance for robust spin-photon entanglement schemes. Demonstrating acoustic control of this centre would therefore represent an important step toward hybrid quantum systems in which optical, spin, and mechanical degrees of freedom are integrated on a single semiconductor platform. Despite this promise, efficient SAW resonators on 6H-SiC and SAW-driven ODMR of V3 centres have not yet been established to the same extent as related systems in 4H-SiC. This defines the central research gap addressed in this thesis: the implementation and characterization of acoustic spin manipulation for V_{Si}^- -related V3 centres in the 6H-SiC polytype.

1.1.2 Aim of this work

The aim of this thesis is to investigate acoustic control of spin qubits in 6H-SiC by using surface acoustic waves to manipulate the spin dynamics of V3 centers. The work focuses on the design, fabrication, and characterization of SAW devices on 6H-SiC substrates, followed by optical and spin-resonance measurements of the V3 center under acoustic excitation.

The first objective is the fabrication of SAW resonators suitable for operation on 6H-SiC. This involves the deposition of piezoelectric thin film (ZnO) and the patterning of interdigital transducers (IDTs) for the excitation and detection of SAWs. The fabricated acoustic devices are then characterized by radio-frequency measurements of their reflection and transmission coefficients in order to determine their resonance frequencies, acoustic response, and device performance. Where applicable, spatial mapping of the acoustic fields using interferometry based techniques provides additional information about the SAW propagation and confinement.

The second objective is to perform optical characterization of the V3 centres and to measure SAW-driven ODMR under suitable magnetic-field conditions. In particular, transverse magnetic fields are used to enable and study acoustically driven spin transitions. By comparing the optical response with the applied acoustic excitation, the thesis aims to identify signatures of SAW-induced spin manipulation and to evaluate the efficiency of the spin-phonon coupling.

Overall, this work seeks to demonstrate efficient SAW resonators on 6H-SiC, realize acoustic spin control of V3 centres, and provide a detailed analysis of the spin-phonon interaction in both the ground and excited states of the defect. Achieving these goals would contribute to the development of mechanically controlled spin qubits in SiC and support the broader effort to build integrated hybrid quantum devices based on optically active defects, phonons, and photons.

Chapter 2

Literature Review

2.1 Silicon Carbide as a Quantum Material

2.1.1 Crystal structure and polytypism

Silicon Carbide is a large bandgap semiconductor compound consisting of Silicon and Carbon. It consists of three crystalline polymorphs. The cubic (β form) and the hexagonal (α form)[17] and the rhombohedral form[18]. These names describe the form and the shape structure the compounds follow.

After its first wide-scale production was initiated by Edward Goodrich Acheson [19] in 1891, over 250 different polytypes of SiC was reported by 2006[20]. Although there are lots of α form structures, there is only a single polytype of the β namely, 3C-SiC.

In the naming scheme of SiC polytypes, H represents hexagonal, and C represents cubic structure. The number coming before these letters represents the periodicity of the stacking. A shorthand has been developed to describe the almost infinite number of possible SiC polytype crystal structures. Each SiC bilayer, whilst keeping the tetrahedral bonding scheme of the crystal, can be placed in one of three possible positions with respect to the lattice. These positions are generally represented by three reciprocal figures of A, B and C. A and B do not change the direction of the layer or twist it, which does not change the lattice. C, however, changes the lattice. Depending on the stacking order, the bonding between Si and C atoms in adjacent bilayer planes can have either zinc-blende, cubic, or wurtzite, hexagonal, nature. Zinc-blende bonds are rotated 60° with respect to nearest neighbors, while hexagonal bonds are mirror images. By the various combinations of these ABC structures, various SiC polytypes are formed[21]. For example, 3C-SiC follows ABC structure, 2H-SiC follows ABAB, 4H-SiC follows ABCB, and 6H-SiC follows ABCACB structure, as can be seen in Fig. 2.1.

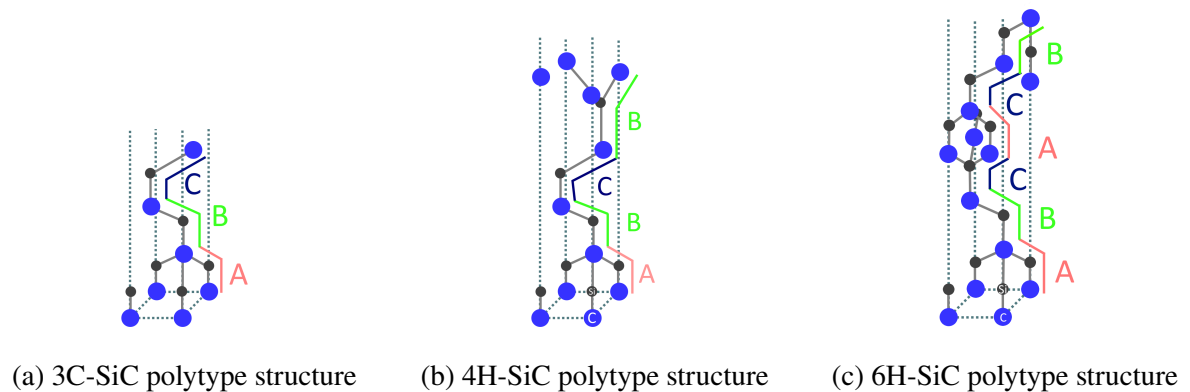


Figure 2.1: Crystal structures of the 3C-SiC, 4H-SiC, and 6H-SiC polytypes.

2.1.2 Properties and Limitations of 3C-SiC Compared with 4H- and 6H-SiC

The three technologically important SiC polytypes considered here are 3C-SiC, 4H-SiC, and 6H-SiC. They have the same chemical composition, but differ in the stacking sequence of Si–C bilayers. This polytypism gives rise to different crystal symmetries and material properties: 3C-SiC is cubic, whereas 4H-SiC and 6H-SiC are hexagonal [6, 22]. In the comparative review by Capan, the band gaps are given as 2.36 eV for 3C-SiC, 3.26 eV for 4H-SiC, and 3.02 eV for 6H-SiC. The same comparison also gives a lower critical electric field for 3C-SiC than for the two hexagonal polytypes [23]. These differences are important because a wider band gap and a larger breakdown field are favourable for high-power, high-temperature, and high-voltage operation.

A major advantage of 3C-SiC is its compatibility with silicon substrates. Since 3C-SiC can be grown epitaxially on large-area silicon wafers, it offers a route toward lower-cost fabrication and integration with established silicon technology. This makes 3C-SiC attractive for applications such as MEMS (Micro-Electro-Mechanical Systems), optical devices, and biomedical devices, especially when suspended structures are required, because the silicon substrate can be selectively removed more easily than a bulk SiC substrate [22]. However, this advantage is accompanied by a central materials problem: the lattice and thermal-expansion mismatch between 3C-SiC and Si can introduce defects at or near the 3C-SiC/Si interface. Interface-related electrically active defects in heteroepitaxial 3C-SiC grown on Si were investigated by Kato *et al.* [24].

The smaller band gap of 3C-SiC also limits its usefulness in high-power and high-temperature devices when compared with 4H-SiC and 6H-SiC. Although 3C-SiC has been studied for electronic applications because of its low-cost growth route, Capan emphasizes that its lower band gap and lower critical electric field are disadvantages relative to the hexagonal polytypes [23]. In addition, defect studies in 3C-SiC are less mature. The review summarizes that several deep-level defects have been reported in 3C-SiC, but that early studies could not always clearly distinguish whether these levels originated from the growth process, the Si/3C-SiC interface, or intrinsic defects in 3C-SiC itself. Later work connected an important deep level in n-type 3C-SiC with the carbon vacancy, but the nature, stability, and device impact of electrically active defects in 3C-SiC remain open questions [23].

In the 4H polytype of SiC, the stacking sequence gives rise to two crystallographically nonequivalent silicon sites, commonly denoted as the quasihexagonal (*h*) and quasicubic (*k*) sites. Silicon vacancies can therefore occur in two different local environments, leading to the V1 and V2 silicon-vacancy-related centres. These centres are generally associated with negatively charged silicon vacancies, V_{Si}^- , with spin $S = 3/2$. Ivády *et al.* identify the V1

centre with V_{Si}^- at the h site and the V2 centre with V_{Si}^- at the k site in 4H-SiC [15]. Due to their different local crystal environments, the two centres exhibit different optical and spin properties. In particular, the V2 centre is important for room-temperature spin-based applications, including magnetometry and related quantum-sensing schemes [15].

Beyond these defect-specific properties, 4H-SiC has reached a higher level of technological maturity. This polytype has become the dominant platform for power electronics and has benefited from major improvements in wafer quality, epitaxial growth, and processing technology [6, 22]. The maturity of 4H-SiC is important for quantum devices as well, because quantum applications require reproducible material quality and controlled defect formation. Silicon-vacancy defects in 4H-SiC have been studied extensively, including their charge-state identification and control [25]. Single-spin coherent control in SiC at room temperature has also been demonstrated [26], and optically active spin defects in ultrapure SiC have been engineered as near-infrared single-photon emitters [27]. These results make 4H-SiC a particularly strong candidate for spin-based and photonic quantum technologies.

In the 6H polytype of SiC, which has 33% hexagonality, the stacking sequence gives rise to three crystallographically nonequivalent silicon sites: one quasihexagonal site, denoted h , and two quasicubic sites, denoted k_1 and k_2 . Silicon vacancies can therefore occur in three different local environments, giving rise to the V_1 , V_2 , and V_3 silicon-vacancy-related centers. The h site corresponds to V_1 , while the k_1 and k_2 sites correspond to V_3 and V_2 , respectively [28]. As we have discussed, in 6H-SiC, the close-packed hexagonal bilayers follow the stacking sequence ABCACB. The local (h/k) site notation is determined by comparing the two neighbouring bilayers above and below a given bilayer. If these neighbouring bilayers occupy the same stacking position in the ABC notation, the central bilayer is classified as an (h) site, corresponding to a hexagonal local environment. If the neighbouring bilayers occupy different stacking positions, the central bilayer is classified as a (k) site, corresponding to a cubic-like local environment. For the ABCACB stacking sequence of 6H-SiC, the (A) bilayers are sandwiched between equivalent neighboring bilayers and are therefore labeled (h), whereas the (B) and (C) bilayers have inequivalent neighboring bilayers and are labeled as the two distinct cubic-like sites, (k_1) and (k_2), respectively [29]. With these all settled we can visualise the sites of the colour centres as in Fig. 2.2.

The 6H-SiC polytype is less dominant than 4H-SiC in power electronics, mainly because of its lower electron mobility, but it still has a wide band gap, high thermal conductivity, and strong robustness under demanding conditions [23]. Its relevance for quantum technologies has also increased because point-defect colour centres in 6H-SiC have been investigated using photoluminescence spectroscopy. Ousdal *et al.* studied the formation and thermal stability of such defects in 6H-SiC and reported defect-related emissions associated with colour-centre

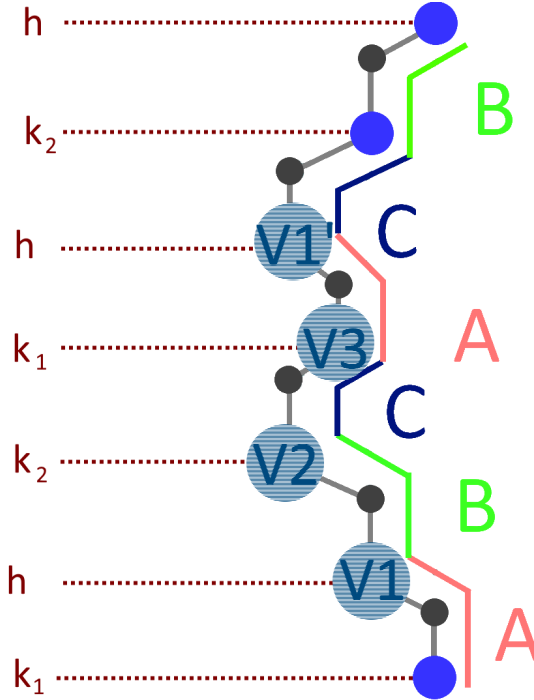


Figure 2.2: Schematic representation of the inequivalent silicon-vacancy configurations in 6H-SiC along the c -axis. The h , k_1 , and k_2 labels denote the local environments of the silicon sites. A silicon vacancy on an h site gives the V_1 center, while vacancies on k_1 and k_2 sites give the V_3 and V_2 centres, respectively. The V_1' label denotes an additional optical feature associated with the h -site silicon vacancy, rather than a distinct crystallographic site.

candidates such as silicon vacancies, divacancies, and carbon antisite-vacancy pairs [30]. This indicates that 6H-SiC may also provide a useful platform for quantum applications, although the coherence properties of its defects are not yet as extensively developed as those of 4H-SiC.

For 3C-SiC, quantum applications have also been explored. Falk *et al.* showed that 3C-, 4H-, and 6H-SiC can host coherent, optically addressable defect spin states, including room-temperature quantum coherence [31]. Calusine *et al.* later demonstrated 3C-SiC photonic crystal cavities with integrated colour centres and discussed their potential for wafer-scale quantum information and sensing devices [32]. However, La Via *et al.* note that further development of 3C-SiC applications is still limited by material-process maturity and the lack of dedicated foundry infrastructure for these fields [22]. Therefore, while 3C-SiC is promising for scalable and Si-compatible device architectures, it is not yet as established as 4H-SiC for controlled quantum-device fabrication.

Overall, 3C-SiC is attractive because it can be integrated with silicon substrates and can support lower-cost, wafer-scale device concepts. Its disadvantages are the smaller band gap,

lower critical electric field, interface-related defect formation, and less mature defect characterization. In contrast, 4H-SiC combines greater technological maturity with well-studied optically addressable spin defects, making it the most developed SiC platform for quantum technology devices. The 6H-SiC polytype is also promising because its defect-related optical behaviour may support future spin-qubit and single-photon-emission applications. For this reason, 4H-SiC and 6H-SiC are treated here as more suitable and better-supported material platforms for the quantum-technology devices considered in this thesis, while 3C-SiC remains relevant for Si-compatible and wafer-scale device concepts.

2.1.3 6H-SiC Inverted Fine Structure

The discussion in this section follows the analysis of Breev *et al.* [1]. The key feature of the V3 silicon-vacancy centre in 6H-SiC is the inversion of its excited-state fine structure. For the V2 centre, and approximately for the V1/V1' system, the lowest optically relevant excited state has predominantly 4A_2 character. In the V3 centre, this order is reversed: the 4E multiplet lies below the 4A_2 multiplet. Thus, after optical excitation and relaxation within the excited-state manifold, emission occurs mainly from a state with different orbital symmetry than in the conventional case.

This reversal is important because the symmetry of the lowest excited state determines the optical selection rules. Transitions from a 4A_2 excited state to the 4A_2 ground state are mainly associated with dipole components parallel to the crystal c -axis. By contrast, transitions from a 4E excited state involve dipole components predominantly perpendicular to the c -axis. Therefore, the inverted ordering in V3 does not merely shift the excited-state energies; it changes which optical transition dominates and consequently changes the polarization character and emission geometry of the centre.

The practical advantage is that dipoles oriented perpendicular to the c -axis preferentially radiate along the c -axis. Since planar SiC photonic structures are typically accessed from the growth surface, this emission geometry is favourable for out-of-plane photon extraction. The V3 centre is therefore better matched to planar photonic devices than centres whose dominant transition is associated with dipoles along the c -axis. In addition to improving photon collection, the altered selection rules are also relevant for spin–photon interfaces: depending on the relative strength of spin–orbit interaction and local deformation, the V3 centre can support optical transitions that couple spin states to circularly or linearly polarized photons. This makes the inverted fine structure useful not only for emission directionality, but also for proposed spin–photon entanglement schemes.

Breev *et al.* [1] also report distinct zero-field splittings for the spin of the 4A_2 in V2 and V3 centres in 6H-SiC. In the ground state, the values are $2D = -28$ MHz for V3 and

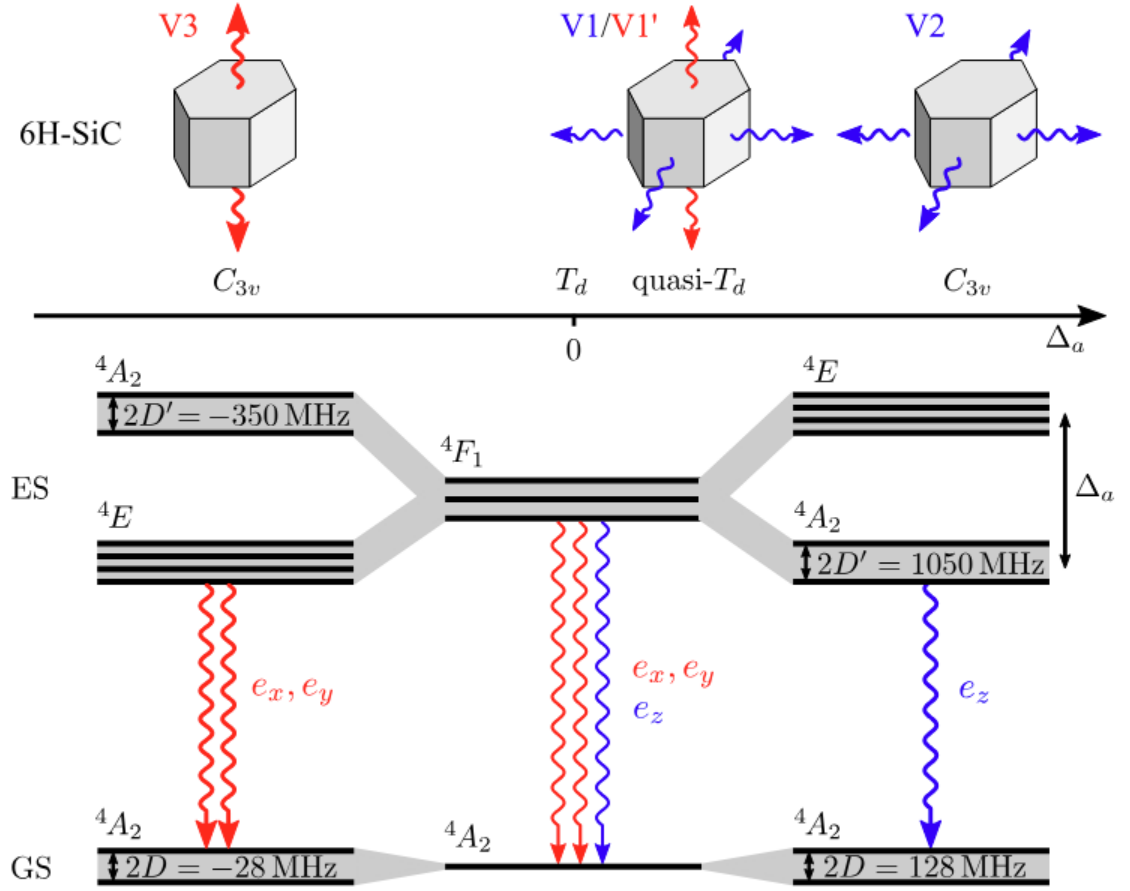


Figure 2.3: Schematic representation of the ground-state (GS) and excited-state (ES) multiplets of the silicon-vacancy (V_{Si}) centres in 6H-SiC. The polarization-dependent directional emission is associated with the ES structure. The zero-field splittings of the GS and ES 4A_2 quartets of the V_2 and V_3 centres were determined experimentally. Adapted from Breev et al. [1].

$2D = 128$ MHz for V_2 . In the excited state, they report $2D' = -350$ MHz for V_3 and $2D' = 1050$ MHz for V_2 . These opposite signs and different magnitudes show that the V_3 centre has a spin structure that is clearly distinct from V_2 in both the ground and excited states. Together with the inverted ordering of the excited-state multiplets, these parameters identify V_3 as a silicon-vacancy centre with a distinct electronic and spin configuration rather than simply a spectrally shifted counterpart of V_2 .

2.2 Spin Qubits and Spin Qudits in SiC

In classical computing we use the binary system, which is composed of ones (1) and zeros (0), to store and process data. In quantum computing, the corresponding two-level system can also exist in a superposition. In such a state, a quantum bit, or qubit, can be described as a linear combination of the states $|0\rangle$ and $|1\rangle$. Calculations in quantum computing are therefore carried out using the probability amplitudes of these states.

$$|\psi\rangle = \alpha |0\rangle + \beta |1\rangle \quad (2.1)$$

In Eq. 2.1, $|\psi\rangle$ is the quantum state of the qubit, while α and β are complex probability amplitudes. By changing these amplitudes, one can represent the full range of qubit states between $|0\rangle$ and $|1\rangle$.

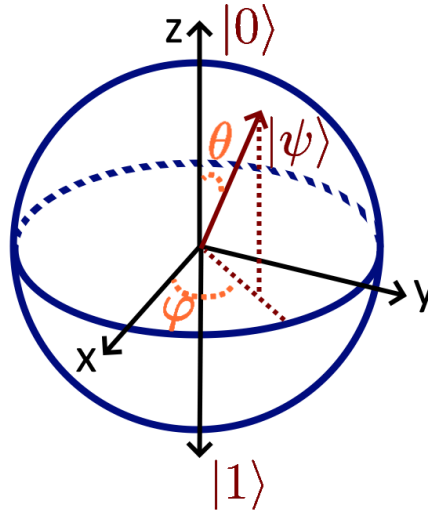


Figure 2.4: Representation of a Bloch sphere.

According to the Born rule, the probability of measuring the state $|0\rangle$ is $|\alpha|^2$, while the probability of measuring the state $|1\rangle$ is $|\beta|^2$. Therefore, for a normalized qubit state,

$$|\alpha|^2 + |\beta|^2 = 1. \quad (2.2)$$

The amplitudes can be parametrized as

$$\alpha = \cos\left(\frac{\theta}{2}\right) \quad (2.3)$$

and

$$\beta = e^{i\phi} \sin\left(\frac{\theta}{2}\right), \quad (2.4)$$

With this parametrization, a single qubit can be represented geometrically on a sphere, called the Bloch sphere, as shown in Fig. 2.4.

The term qubit is coined for a two-level quantum system, whereas the term qudit denotes a quantum system with d levels [33].

Physically, a qubit can be realised using electron spins, nuclear spins, photon polarization, or charge states in superconducting Josephson-junction systems [34]. In the following section, we discuss how such spin states arise and how they can be used to realise qubits and qudits.

2.2.1 Spin states and zero-field splitting

The isolated silicon vacancy (V_{Si}) is a fundamental intrinsic defect in SiC. Because SiC polytypes contain inequivalent silicon lattice positions, different forms of V_{Si} can occur. In 4H-SiC, there are two silicon vacancy sites: one hexagonal site, denoted as h , and one quasi-cubic site, denoted as k . In 6H-SiC, three silicon vacancy sites are present: one hexagonal site, h , and two quasi-cubic sites, k_1 and k_2 . Each of these vacancies can also appear in several charge states within the band gap [5].

Atomic-scale defects in SiC are promising solid-state spin systems because they can be optically addressed and controlled using radio-frequency fields. Kraus *et al.* emphasize that these defects are attractive for sensing applications due to the technological maturity of SiC, their near-infrared optical activity, and their compatibility with MHz radio-frequency control fields [9].

In 6H-SiC, several intrinsic defects can give rise to optically detected magnetic resonance (ODMR) signals. The primary stoichiometric defects in solids include Schottky and Frenkel defects. In SiC, a divacancy $V_{\text{Si}} - V_{\text{C}}$, formed from a chemically bound silicon vacancy and carbon vacancy, is an example of a Schottky defect. Other vacancy-related configurations, such as $V_{\text{Si}}(V2)$ and $V_{\text{Si}}(V3)$, are also observed in 6H-SiC. Frenkel defects can also occur, for example when a silicon vacancy is accompanied by a silicon interstitial atom, forming a $V_{\text{Si}} - Si_i$ pair [9].

A common feature of these intrinsic defects in SiC is that they possess a high-spin ground state. In particular, Kraus *et al.* describe the relevant centres in 6H-SiC using a spin Hamiltonian for half-integer spin centres with total spin $S = 3/2$ [9]. For such systems, the spin sublevels are labelled by

$$m_s = \pm\frac{3}{2}, \pm\frac{1}{2}.$$

For spin systems with $S > 1/2$, interactions with the local crystal field can split the spin sublevels even when no external magnetic field is applied. This effect is known as zero-field splitting. At zero magnetic field, the crystal-field interaction splits the $m_s = \pm 3/2$ and $m_s = \pm 1/2$ doublets. For $S = 3/2$, the zero-field splitting between these two doublets is $2D$. This spin-3/2 structure is the starting point for constructing the effective spin Hamiltonian used to describe the Zeeman interaction, zero-field splitting, and strain-induced transitions in SiC defects [9].

Detailed derivation of these equations which forms the heart of our simulations and calculations can be seen on Appendix A.

Static Spin Hamiltonian

Next, we consider the unperturbed static Hamiltonian, i.e., the Hamiltonian in the absence of the SAW drive. It contains the zero-field splitting term and the Zeeman interaction with an in-plane magnetic field:

$$\hat{H}_0(\phi_B) = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + g\mu_B \vec{B}_{\text{DC}}(\phi_B) \cdot \vec{S} \quad (2.5)$$

Here, D is the zero-field splitting parameter, S is the total spin, g is the effective Landé factor, μ_B is the Bohr magneton, and $\vec{S} = (S_x, S_y, S_z)$ is the spin operator. The static magnetic field is assumed to lie in the xy plane. For a field magnitude B and in-plane angle ϕ_B , the magnetic field is

$$\vec{B}_{\text{DC}}(\phi_B) = B(\cos \phi_B \hat{x} + \sin \phi_B \hat{y}) \quad (2.6)$$

Substituting Eq. (2.6) into Eq. (2.5) gives

$$\hat{H}_0(\phi_B) = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + g\mu_B B(\cos \phi_B S_x + \sin \phi_B S_y) \quad (2.7)$$

This Hamiltonian defines the unperturbed spin system. The SAW-induced strain interaction is not included in $H_0(\phi_B)$, because it is treated as a time-dependent external perturbation. The term proportional to $S(S+1)$ is a constant energy offset for fixed spin S and therefore the energy difference between the eigenstates does not get affected.

For each magnetic-field angle, the eigenvalue problem is

$$\hat{H}_0(\phi_B) |i(\phi_B)\rangle = E_i(\phi_B) |i(\phi_B)\rangle \quad (2.8)$$

For a spin-3/2 system, the Hilbert space is four-dimensional, giving the angle-dependent

eigenbasis

$$\{|1(\phi_B)\rangle, |2(\phi_B)\rangle, |3(\phi_B)\rangle, |4(\phi_B)\rangle\} \quad (2.9)$$

These states depend on ϕ_B because the direction of the Zeeman term changes as the in-plane magnetic field is rotated.

2.3 Acoustic Waves and Spin-Acoustic Coupling

2.3.1 Surface Acoustic Waves

The first research on the topic of surface acoustic waves can be traced back to Lord Rayleigh, to his paper "On Waves Propagated along the Plane Surface of an Elastic Solid" dated 1886[35]. Lord Rayleigh first suggested that the surface waves described in his work could be significant in both earthquake phenomena and the impact of elastic solids. Although Rayleigh-wave theory was later taken up in geophysical studies, earthquake surface waves in layered media are more accurately represented by Love waves, a shear-horizontal class of surface acoustic waves that propagate along stratified structures [36].

Several types of surface acoustic waves can exist depending on the material system and wave polarization, including Love waves in layered structures, shear waves, and surface-skimming bulk waves. The wave originally described by Rayleigh is now known as the Rayleigh wave when it propagates in an isotropic medium. Rayleigh waves are the fundamental type of surface acoustic wave and are confined near the surface of the material. Their displacement amplitude is largest at the surface and decreases approximately exponentially with depth, over a distance on the order of one wavelength. In an isotropic medium, Rayleigh waves contain both horizontal and vertical displacement components, giving rise to elliptical particle motion. As a result, Rayleigh waves are often treated as quasi-two-dimensional waves. Their phase velocity is typically lower than that of bulk acoustic waves in the same medium, approximately 10% lower than the bulk shear-wave velocity and about 50% lower than the bulk longitudinal-wave velocity. This lower velocity prevents efficient coupling into bulk acoustic modes, so the acoustic energy remains confined near the surface rather than radiating into the bulk [36].

Some well-known materials which show such elasticity and piezoelectric behaviour are ZnO [37], $LiNbO_3$ [38], and AlN [39]. With their piezoelectric properties, such substrates are ideal materials for SAW generation and detection[37]. SAW devices are usually layered structures where a piezo-material is disposed on top of a supporting substrate[40]. The sub-layer substrate(s) can vary depending on the specific application. Some examples are Sapphire $\alpha - Al_2O_3$ [41] and $4H-SiC$ [14]. These materials are usually selected for their

semiconductive properties.

In layered SAW devices, higher-order modes can also appear when the thickness of the piezoelectric layer becomes comparable to the SAW wavelength. Under these conditions, the wave phase velocity and electromechanical coupling coefficient become frequency-dependent, leading to dispersive device behaviour. One important higher-order mode is the Sezawa mode, which was introduced by Sezawa and Kanai in 1935 in their study of discontinuities in Rayleigh-wave dispersion curves[42]. In SAW devices, Sezawa waves can be generated in layered structures when the upper piezoelectric layer has a lower acoustic phase velocity than the underlying substrate. This mode appears at higher frequencies in addition to the fundamental Rayleigh mode and, like Rayleigh waves, is excited through the piezoelectric effect by applying a harmonic electrical source [43].

Surface acoustic waves (SAWs) provide an attractive alternative to conventional microwave (MW) driving for controlling spin interactions in defects such as silicon-vacancies and divacancies in SiC[16][39]. In 4H-SiC, the V2 center can be efficiently addressed by MW-driven ODMR at room temperature, but MW excitation is restricted to magnetic-dipole-allowed transitions with $\Delta m_S = \pm 1$. By contrast, dynamic strain produced by acoustic fields can drive both $\Delta m_S = \pm 1$ and $\Delta m_S = \pm 2$ spin transitions, including transitions in the ground and excited state manifolds. This capability is particularly important because SAW-driven spin resonances have been shown to manipulate both V1 and V2 silicon-vacancy centres in 4H-SiC up to room temperature[13], whereas MW-driven control of the V1 centre has been limited to cryogenic conditions[44]. These results motivate the use of SAWs as a strain-mediated control mechanism for silicon-vacancy centers in 6H-SiC, where multiple inequivalent vacancy configurations are present.

2.3.2 Strain fields generated by acoustic waves

Interdigital Transducers - IDTs

Interdigital Transducers (IDTs) are sets of dovetailed fringes, settled on a piezoelectric crystal. Usually two pairs of these IDTs are used together as one being the transmitter and the latter being the receiver. It was first realised by White and Volmer in 1965 [45]. This device was utilised to create band-devices such as pass filters, oscillators and, space charge coupling was also demonstrated by 1979[40].

The common waves which are induced by IDTs could be grouped in two; bulk and surface acoustic waves[2].

In contrast to bulk acoustic waves (BAWs), SAWs are confined to the surface of a material and propagate as guided waves. For a point source, their amplitude decreases proportionally

to $1/\sqrt{r}$, whereas excitation by a line source, such as an IDT, results in significantly reduced attenuation. This property makes SAWs well suited for signal and power transmission. However, their generation typically requires more restrictive conditions than that of bulk waves, such as the aperture, wave propagation direction, number of the IDT fingers etc. as can be seen on 2.5 [2]. IDTs can come in various shapes and size. Song et. al. [2]

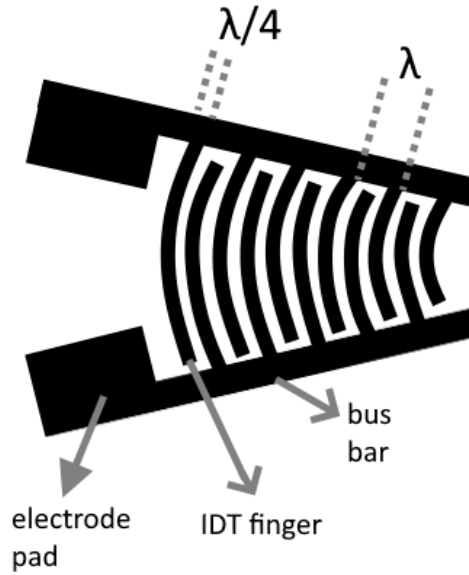


Figure 2.5: Componenets of an IDT

gives eight different shapes. To simplify the terms we can roughly divide them into two groups: toroidal and linear IDTs. Basically for the linear IDTs, the difference in design can come from the number of fingers on each side, thickness, length, angle and the curvature of the fingers. Different IDT configurations can be used depending on the desired acoustic response; representative examples, drawn with reference to the configurations discussed by Song et al. [2], are shown in Fig. 2.6.



Figure 2.6: Examples of IDT configurations. The illustrations were created by the author based on the configurations described by Song et al. [2].

As also have been demonstrated by [14] and [39], on our setup we will also be using a similar focus IDT. As we are interested in observing the spin properties of a small cluster of

vacancies, a focus IDT enables us to narrow down the region of inspection whilst increasing the signal strength.

2.3.3 Spin-Phonon Coupling Model

This subsection presents the spin-phonon coupling model used to calculate SAW driven transition strengths as a function of the in-plane magnetic field angle ϕ_B . In this model, the SAW is treated as a coherent acoustic phonon mode that produces a time-dependent strain field. The strain field couples to the spin degrees of freedom and drives transitions between the eigenstates of the static spin Hamiltonian. As also demonstrated by Hernández-Mínguez et. al [14], the IDTs couple to Rayleigh SAW modes within their resonance band.

The calculation proceeds in three steps. First, the strain induced spin-phonon coupling operator is defined for the relevant SAW geometry. Second, the static Hamiltonian is constructed and diagonalized for each value of ϕ_B . Third, transition matrix elements of this operator are evaluated in the angle dependent eigenbasis of the static Hamiltonian. The squared modulus of these matrix elements gives the transition intensity.

Travelling-Wave SAW Strain

The SAW produces a time-dependent strain field that couples to the spin degrees of freedom. The strain tensor is defined from the displacement field $\vec{u}$ as

$$\epsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right). \quad (2.10)$$

For a travelling surface acoustic wave (SAW), the strain field depends on both position and time. In complex notation, it can be written as

$$\epsilon_{ij}(x, t) = \Re \left[\epsilon_{ij}^{(0)} e^{i(kx - \omega t)} \right], \quad (2.11)$$

where $\epsilon_{ij}^{(0)}$ is the complex strain amplitude. The wave number k is given by

$$k = \frac{2\pi}{\lambda}, \quad (2.12)$$

where λ is the wavelength. The angular frequency ω is given by

$$\omega = 2\pi f, \quad (2.13)$$

where f is the frequency.

The phase of each strain component is contained in its complex amplitude. The longitudinal strain components are chosen real, while the shear component is chosen imaginary so that it is phase-shifted by $\pi/2$:

$$\epsilon_{xx}^{(0)} = u_{xx}, \quad \epsilon_{zz}^{(0)} = u_{zz}, \quad \epsilon_{xz}^{(0)} = i u_{xz}. \quad (2.14)$$

Using Eq. (2.11), the xx strain component becomes

$$\epsilon_{xx}(x, t) = \Re[u_{xx} e^{i(kx - \omega t)}] = u_{xx} \cos(kx - \omega t), \quad (2.15)$$

and similarly,

$$\epsilon_{zz}(x, t) = \Re[u_{zz} e^{i(kx - \omega t)}] = u_{zz} \cos(kx - \omega t). \quad (2.16)$$

For the shear component,

$$\begin{aligned} \epsilon_{xz}(x, t) &= \Re[i u_{xz} e^{i(kx - \omega t)}] \\ &= \Re[i u_{xz} \cos(kx - \omega t) - u_{xz} \sin(kx - \omega t)] \\ &= -u_{xz} \sin(kx - \omega t). \end{aligned} \quad (2.17)$$

Thus, the shear strain is phase-shifted by $\pi/2$ relative to the normal strain components, with the sign fixed by the convention $\epsilon_{xz}^{(0)} = i u_{xz}$ and $e^{i(kx - \omega t)}$.

The time- and position-dependent SAW driving perturbed Hamiltonian is

$$\hat{H}_{\text{SAW}}(t, x) = a [\epsilon_{xx}(x, t) S_x^2 + \epsilon_{zz}(x, t) S_z^2 + \epsilon_{xz}(x, t) \frac{1}{2} (S_x S_z + S_z S_x)]. \quad (2.18)$$

where a is the coupling constant.

Substituting the explicit real strain fields gives

$$\begin{aligned} O_{\text{travelling}}(t, x) &= a \left[u_{xx} \cos(kx - \omega t) S_x^2 + u_{zz} \cos(kx - \omega t) S_z^2 \right. \\ &\quad \left. - u_{xz} \sin(kx - \omega t) \frac{1}{2} (S_x S_z + S_z S_x) \right]. \end{aligned} \quad (2.19)$$

For transition matrix elements in the complex-amplitude description, the operator used is

$$\hat{O}_{\text{travelling}}(t, x) = a \left[\epsilon_{xx}^{(0)} S_x^2 + \epsilon_{zz}^{(0)} S_z^2 + \epsilon_{xz}^{(0)} \frac{1}{2} (S_x S_z + S_z S_x) \right]. \quad (2.20)$$

Using Eq. (2.14), this becomes

$$\hat{O}_{\text{travelling}}(t, x) = a \left[u_{xx} S_x^2 + u_{zz} S_z^2 + i u_{xz} \frac{1}{2} (S_x S_z + S_z S_x) \right]. \quad (2.21)$$

Standing-Wave SAW Strain

A standing surface acoustic wave can be represented as two counter-propagating travelling waves with the same magnitude of phase:

$$\epsilon^{(+)}(x, t) = \Re \left[\epsilon_{ij}^{(0)} e^{i(kx + \omega t)} \right] \quad (2.22)$$

and

$$\epsilon^{(-)}(x, t) = \Re \left[\epsilon_{ij}^{(0)} e^{i(kx - \omega t)} \right] \quad (2.23)$$

Their superposition gives the standing wave SW:

$$\begin{aligned} \epsilon_{ij}^{SW}(x, t) &= \epsilon_{ij}^{(+)}(x, t) + \epsilon_{ij}^{(-)}(x, t) \\ &= \Re \left[\epsilon_{ij}^{(0)} \left(e^{i(kx + \omega t)} + e^{i(kx - \omega t)} \right) \right] \\ &= \Re \left[\epsilon_{ij}^{(0)} e^{ikx} \left(e^{i\omega t} + e^{-i\omega t} \right) \right] \\ &= \Re \left[2\epsilon_{ij}^{(0)} e^{ikx} \cos(\omega t) \right] \end{aligned} \quad (2.24)$$

Because the strain tensor components in a SAW have relative phase shifts, the complex amplitude $\epsilon_{ij}^{(0)}$ determines the spatial dependence. The longitudinal and vertical strain amplitudes are purely real, whilst the shear strain amplitude is purely imaginary [2.14](#).

Evaluating the real part for the normal strains gives:

$$\epsilon_{xx}^{SW}(x, t) = 2u_{xx} \cos(kx) \cos(\omega t) \quad (2.25)$$

$$\epsilon_{zz}^{SW}(x, t) = 2u_{zz} \cos(kx) \cos(\omega t) \quad (2.26)$$

Evaluating the real part for the shear strain yields the 90° spatial phase shift:

$$\begin{aligned} \epsilon_{xz}^{SW}(x, t) &= \Re \left[2(-iu_{xz})(\cos(kx) + i \sin(kx)) \cos(\omega t) \right] \\ &= 2u_{xz} \sin(kx) \cos(\omega t) \end{aligned} \quad (2.27)$$

As similar with travelling waves, we can derive the Hamiltonian for the standing waves with eq. [2.18](#).

Substituting the explicit real strain fields gives

$$\begin{aligned} \hat{O}_{\text{standing}}(t, x) &= a \left[2u_{xx} \cos(kx) \cos(\omega t) S_x^2 + 2u_{zz} \cos(kx) \cos(\omega t) S_z^2 \right. \\ &\quad \left. + 2u_{xz} \sin(kx) \cos(\omega t) \frac{1}{2} (S_x S_z + S_z S_x) \right] \end{aligned} \quad (2.28)$$

For transition matrix elements in a standing wave, the harmonic time dependence $\cos(\omega t)$ is factored out, leaving a purely spatial operator:

$$\hat{O}_{\text{standing}}(x) = a \left[\epsilon_{xx}^{\text{SW}}(x) S_x^2 + \epsilon_{zz}^{\text{SW}}(x) S_z^2 + \epsilon_{xz}^{\text{SW}}(x) \frac{1}{2} (S_x S_z + S_z S_x) \right] \quad (2.29)$$

Substituting the explicit spatial amplitudes, this becomes

$$\hat{O}_{\text{standing}}(x) = a \left[2u_{xx} \cos(kx) S_x^2 + 2u_{zz} \cos(kx) S_z^2 + 2u_{xz} \sin(kx) \frac{1}{2} (S_x S_z + S_z S_x) \right] \quad (2.30)$$

Transition Matrix Elements

For a fixed magnetic-field angle ϕ_B , the transition amplitude between an initial state $|i(\phi_B)\rangle$ and a final state $|j(\phi_B)\rangle$ is calculated using the appropriate driving operator $\hat{O}$:

$$M_{ij}(\phi_B) = \langle j(\phi_B) | \hat{O} | i(\phi_B) \rangle \quad (2.31)$$

The corresponding transition intensity is

$$I_{ij}(\phi_B) = |M_{ij}(\phi_B)|^2 \quad (2.32)$$

Equivalently,

$$I_{ij}(\phi_B) = |\langle j(\phi_B) | O | i(\phi_B) \rangle|^2 \quad (2.33)$$

Here, O is chosen according to the SAW geometry:

$$\hat{O} = \begin{cases} \hat{O}_{\text{travelling}}, & \text{travelling SAW} \\ \hat{O}_{\text{standing}}, & \text{standing SAW} \end{cases} \quad (2.34)$$

These intensities are proportional to the square of the corresponding Rabi frequencies and are the quantities used to generate the angular dependence plots. The dependence on ϕ_B enters through the eigenstates $|i(\phi_B)\rangle$ and $|j(\phi_B)\rangle$, while the selected driving operator is fixed by the strain amplitudes, phases, and spin-phonon coupling structure.

Classification of Transitions by Δm_s

To interpret the calculated transition strengths, the transitions are classified using the bare spin-3/2 basis,

$$m_s = -\frac{3}{2}, -\frac{1}{2}, +\frac{1}{2}, +\frac{3}{2} \quad (2.35)$$

The change in spin projection associated with a transition from state i to state j is labeled

by

$$\Delta m_s = m_s(j) - m_s(i) \quad (2.36)$$

The transitions are grouped into two main classes:

- $\Delta m_s = \pm 1$,
- $\Delta m_s = \pm 2$.

These two classes can show different angular patterns because the spin operators entering the strain-driving operator have different tensor structures. For example, if the spin is quantized along the z axis, in the bare spin basis, the relevant operator components have the following dominant selection behaviour:

- S_x^2 mainly contributes to $\Delta m = 0, \pm 2$ transitions,
- S_z^2 contributes to $\Delta m = 0$ terms,
- $S_x S_z + S_z S_x$ contributes to $\Delta m = \pm 1$ transitions.

Therefore, the relative magnitudes and phases of the strain components determine which transition classes are most strongly driven. We can exploit this phenomenon by changing the orientation of the magnetic field, thereby rotating the spin quantization axis relative to the strain field. This changes the contributions of the different spin-operator components to the transition matrix elements and thus allows us to selectively enhance the desired class of transitions. In the travelling-wave case, the phase factor in $\epsilon_{xz}^{(0)} = i u_{xz}$ can modify interference between the longitudinal-strain and shear-strain coupling channels. The observed angular dependence results from the combination of the operator structure, the strain amplitudes, and the field-angle dependence of the eigenstates.

2.3.4 Computational Procedure

For the simulations, the following equations are followed to create the Python code. The complete calculation is performed independently for each magnetic-field angle ϕ_B .

First, the static Hamiltonian is constructed:

$$\hat{H}_0(\phi_B) = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + g\mu_B B (\cos \phi_B S_x + \sin \phi_B S_y) \quad (2.37)$$

Second, the Hamiltonian is diagonalized:

$$\hat{H}_0(\phi_B) |i(\phi_B)\rangle = E_i(\phi_B) |i(\phi_B)\rangle \quad (2.38)$$

Third, the appropriate SAW driving operator is selected from Eq. (2.34). In the simulations this corresponds to using $O_{\text{travelling}}$, or O_{standing} , depending on the chosen SAW geometry.

Finally, the transition intensities are calculated from

$$I_{ij}(\phi_B) = \left| \langle j(\phi_B) | \hat{O} | i(\phi_B) \rangle \right|^2 \quad (2.39)$$

After this calculation is repeated over the full range of ϕ_B , the intensities are grouped according to Δm_s and plotted as functions of the in-plane magnetic-field angle.

Chapter 3

Theoretical Calculations

3.1 Strain

In this section we will start by simulating the equations derived in the section *Spin-Phonon Coupling* 2.3.3 via Python by the procedure described in the section *Computation Procedure* 2.3.4.

Let's start by the Hamiltonian given in the equation (2.37).

$$H_0(\phi_B) = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + g\mu_B B (\cos \phi_B S_x + \sin \phi_B S_y).$$

This is our static Hamiltonian for the 3/2 spin system. The first term describes zero-field splitting, whilst the second term is the Zeeman interaction with the applied magnetic field.

We shall also note that the zero-field splitting parameter D can be temperature dependent. To better give its dynamics, we can use the following equation,

$$D(T) = D_{ref} + \beta(T - T_{ref}) \quad (3.1)$$

where β is the temperature coefficient, D_{ref} is the known value at $T_{ref} = 300K$.

Table 3.1: Temperature dependence of the zero-field splitting in the 4A_2 , GS ($2D$) and ES ($2D'$) on 6H-SiC [1].

Center	$2D$	$2D'$ @ 300 K	β @ 300 K
V2	128 MHz	1050 MHz	-1.16 MHz/K
V3	-28 MHz	-350 MHz	0.84 MHz/K

The temperature shift is obtained as $\beta = 2 dD'/dT$ at room temperature, adapted from the Breev et al.[1].

One must note that although the ZFS of the ES is subject to the temperature coefficient β , the GS is nearly temperature-independent[46]. We will be using the values given in Table 3.1 for our calculations.

Due to the zero-field splitting, the fine-structure levels are already separated at zero magnetic field. Applying an external magnetic field further lifts the degeneracy of the spin sublevels through the Zeeman effect.

The Zeeman effect describes the additional splitting of atomic energy levels in an applied magnetic field. This splitting occurs because the magnetic field couples to the magnetic moment associated with the electron's angular momentum, causing otherwise degenerate magnetic sublevels to acquire different field-dependent energies. In the classical normal Zeeman picture, a transition with zero-field frequency ω_0 with $S = 1/2$ spin, splits into two

components with frequencies ω_0 and $\omega_0 \pm \Omega_L$, where

$$\Omega_L = \frac{g\mu_B B}{\hbar} \quad (3.2)$$

is the Larmor frequency where g is the Landé g -factor, μ_B is the Bohr magneton, B is the magnetic field strength and $\hbar$ is the reduced Planck constant. Therefore, once the fine-structure levels have been separated by zero-field splitting, the applied magnetic field produces an additional field-dependent separation of the spin sublevels [47].

We can see the effects of the Zeeman effect on the spin sublevels shown in Fig. 3.1.

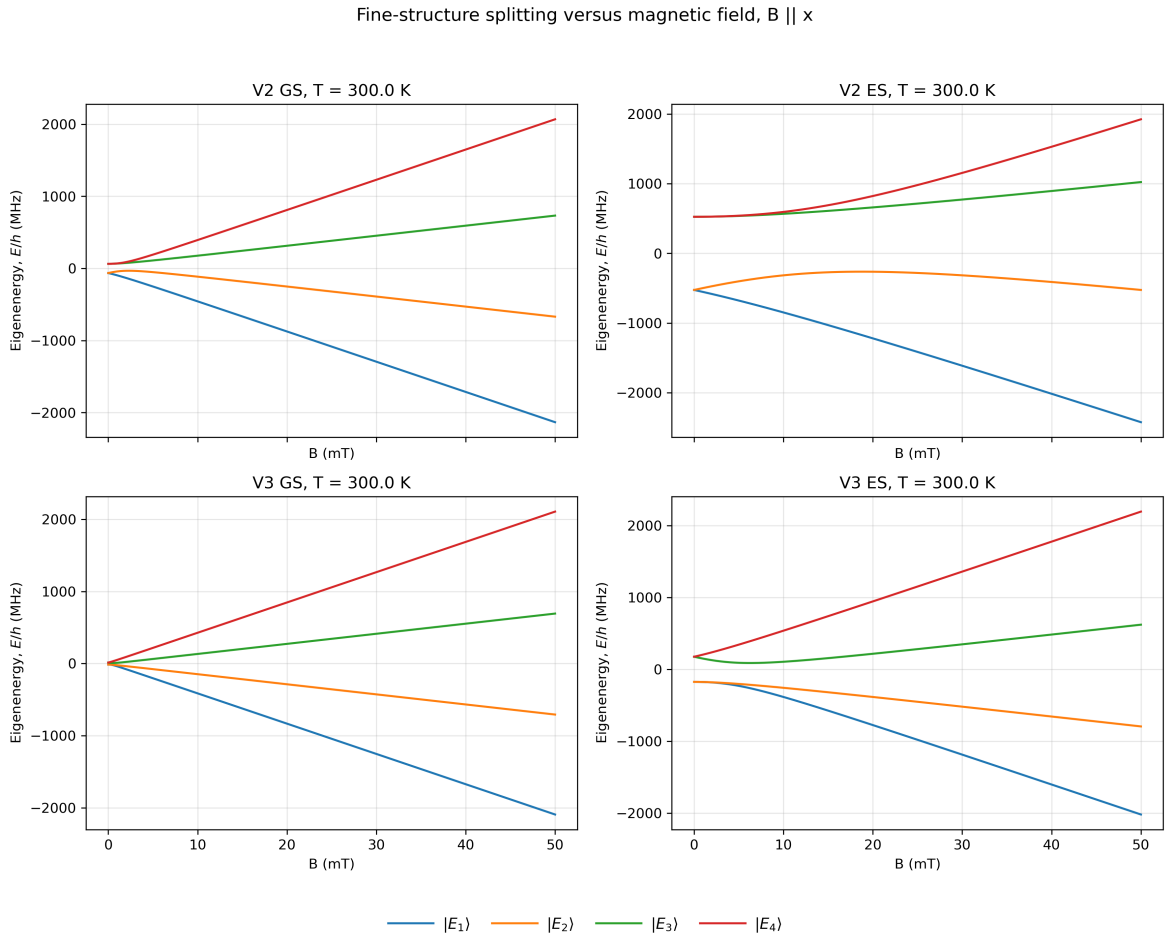


Figure 3.1: Computed energy splittings for a spin-3/2 system under a magnetic field $B \parallel x$ ($\phi_B = 0$) at $T = 300K$. Each curve corresponds to one eigenstate $|E_i\rangle$ obtained by diagonalizing the static spin Hamiltonian H_0/h . The plotted energies are given in MHz which corresponds to the energy divided by Planck's constant

The simulation was run from 0 to 60mT. One can clearly observe the zero-field splitting followed by further diversion as the magnetic field intensity gets higher.

So far we have seen how the 6H-SiC spin levels behaves under varying magnetic field. We shall now examine how the SAWs couple with the system.

For a SAW propagating along x-axis we can subsidize the strain into three components of u_{xx} , u_{zz} and u_{xz} . The SAW can be described as a solution of the elastic equations with boundary conditions such that the amplitude deep into the substrate is 0.

$$u_x(z \rightarrow \infty) = u_x(z \rightarrow \infty) = 0 \quad (3.3)$$

Strain depth profile data was provided by Dr. Hernández-Mínguez.

Figure 3.2 represents the strain values for varying depth. We have a three layered sample. ZnO $0.035 \mu m$, SiO_2 $0.735 \mu m$ followed by 6H-SiC $500 \mu m$.

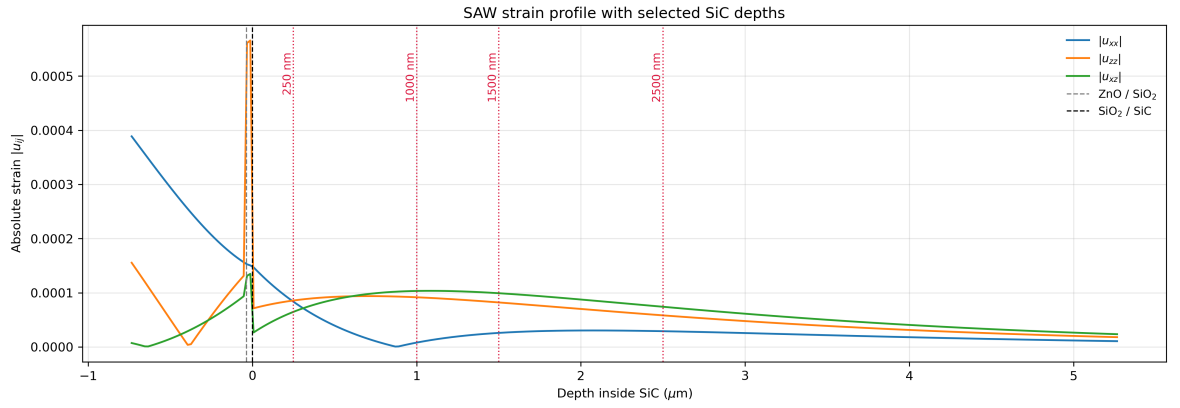


Figure 3.2: SAW strain depth profile showing the strain components u_{xx} , u_{zz} , and u_{xz} as a function of depth. The data were provided by Dr. Hernández-Mínguez, Paul-Drude-Institut für Festkörperelektronik (PDI).

On the x-axis of the 3.2, SiC starts from the $0 \mu m$ point.

Wavelength (nm)	u_{xx}	u_{xy}	u_{zz}
250	$2.92e-4$	$4.82e-5$	$1.56e-5$
870.35	$5.93e-7$	$9.30e-5$	$1.02e-4$
1000	$7.95e-5$	$8.64e-5$	$6.68e-5$
1500	$-8.74e-6$	$9.36e-5$	$9.97e-5$
2500	$-2.93e-5$	$7.56e-5$	$9.32e-5$

Table 3.2: Mean strain components u_{ij} as a function of wavelength

One other information about the strain components is their relative phase. On Fig. 3.3 we have the data for the relative phases of the strain components at 250 nm and 2500 nm.

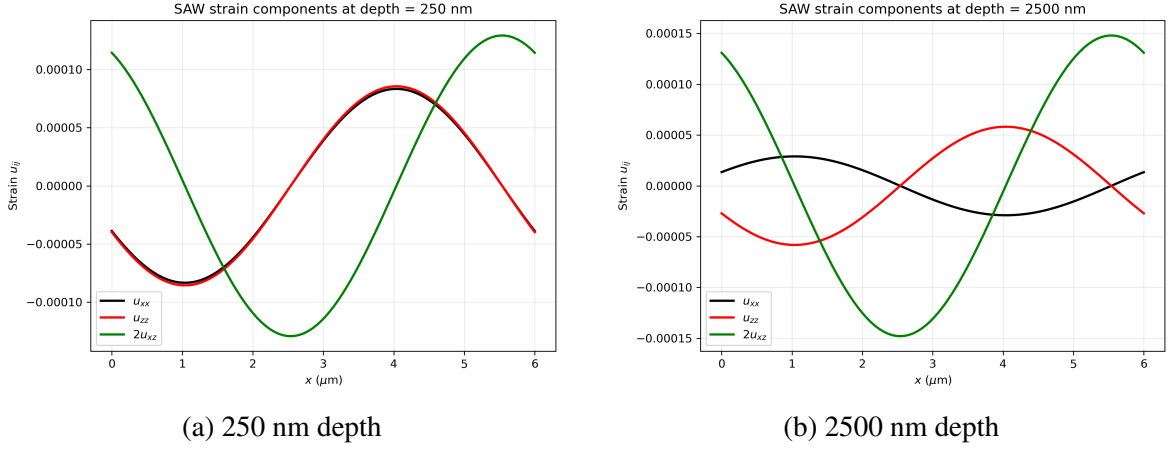


Figure 3.3: SAW strain components as a function of lateral position x at depths of 250 nm and 2500 nm inside SiC. The plotted components are the longitudinal strain u_{xx} , the normal strain u_{zz} , and the engineering shear strain $2u_{xz}$. The data were provided by Dr. Hernández-Mínguez, Paul-Drude-Institut für Festkörperelektronik (PDI).

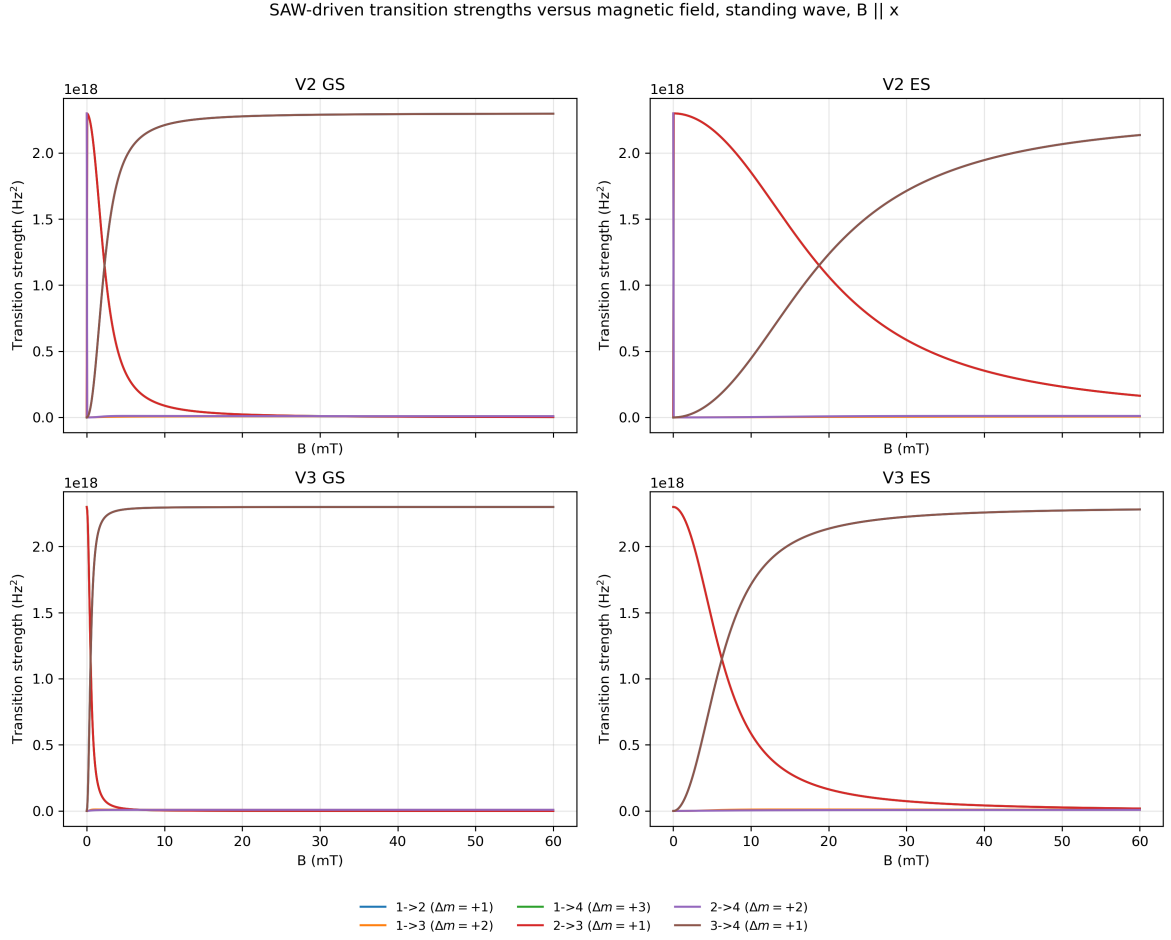
At 250 nm we see that u_{xx} and u_{zz} oscillate in phase and $2u_{xz}$ is 90 degrees out of phase. At 2500 nm we see that u_{xx} and u_{zz} are out of phase by 180 degrees. With the assistance of these graphs we have assigned the sign change of u_{xx} after 1000 nm, shifting from positive to negative amplitude.

Now as we have determined the strain values, we can now introduce the strain operator as in the Eq. 2.39 to see how the specific transition strengths between the sublevels of the fine structure behave under a changing magnetic field.

Fig. 3.4 shows that the magnitude of the magnetic field also affects the magnitude of the transition strengths. Some transitions strengths can be high at some magnetic strengths and diminish at some points.

Although the transition frequencies remain unchanged under in-plane rotation of the magnetic field, Fig. 3.5 shows that the transition strengths vary with the in-plane magnetic-field angle. Later on we can use this phenomenon to selectively induce spin transitions or to identify certain colour centres and transitions ($\Delta m = \pm 1, \Delta m = \pm 2$) in our measurements.

Up until now, we have only examined the case where the magnetic field was applied along the x direction, $B \parallel x$. In Fig. 3.5, we can clearly see the effect of changing the in-plane angle of the magnetic field. As the strain values change for a given depth in the substrate, the transition strengths also change with the angle of the applied magnetic field. For this reason, we have examined various depths in 6H-SiC to have a flexible working environment for our measurements. In Appendix B, one can see the graphs for the V2 GS, V2 ES, V3 GS, and V3 ES angular dependence of the SAW-driven transition strengths as a function of magnetic-field

Figure 3.4: Transition strength versus magnetic field $B \parallel x$.

angle ϕ_B . In this section we have only included the Fig. 3.6 for V3 ES. On these graphs, one can observe $\Delta m = \pm 1$ and $\Delta m = \pm 2$ transitions, and also compare the travelling and standing wave configurations. After examining these graphs, we have decided on creating our V_{Si} colour centres at the depth of 1000 nm. The reason for this is that on Fig. 3.6, one can observe that when the angle between the SAW propagation and the magnetic field is $\phi = 90^\circ$ for Standing SAW configuration, $\Delta m_s = \pm 1$ transition strengths are always zero whilst for the same angle $\Delta m_s = \pm 2$ transitions are the maximum. When we configure the system for $\phi = 0^\circ$ we see that the amplitude of the $\Delta m_s = \pm 1$ transitions are the maximum whilst $\Delta m_s = \pm 2$ are at their minimum. Giving us a distinct advantage in distinguishing between $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions whilst keeping a comparatively high amplitude.

SAWs can be generated either as standing waves or as travelling waves. In our setup, the standing-wave case is the relevant one, because the IDT structure and acoustic reflections can produce two counter-propagating SAWs with the same frequency and similar amplitudes. To

SAW-driven transition strengths versus ϕ_B , standing wave, depth = 1000 nm, $B = 50.0$ mT

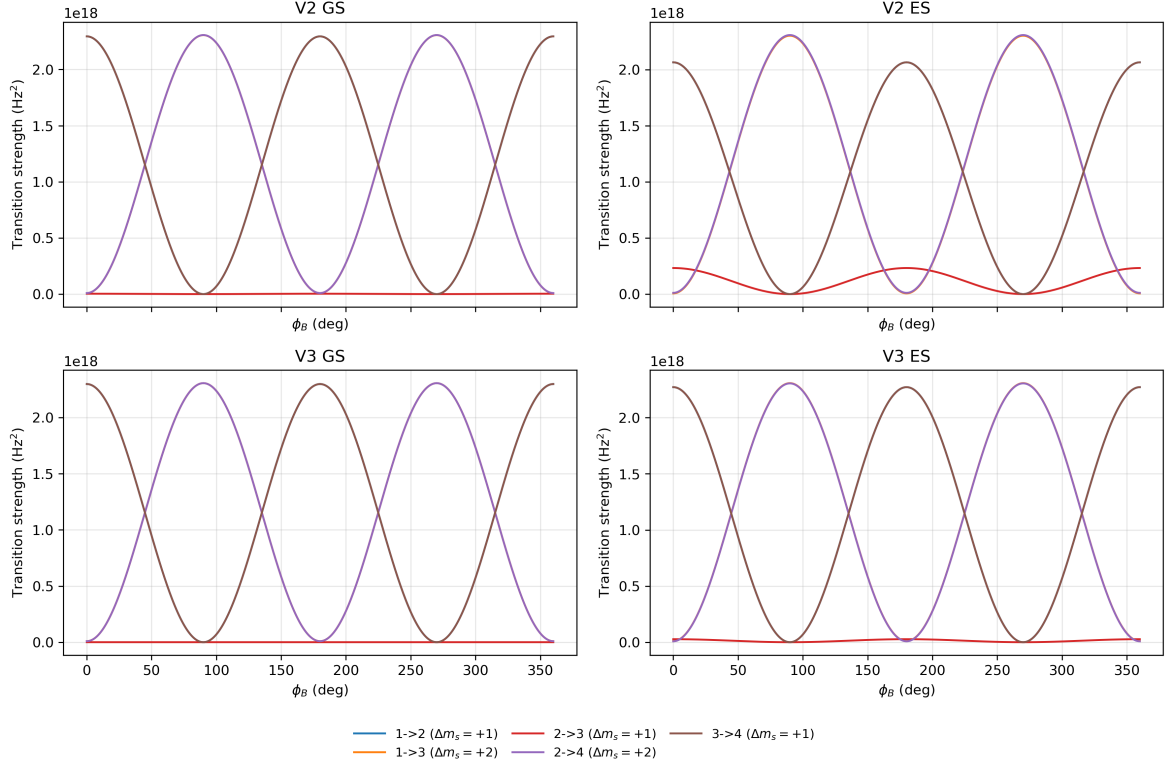


Figure 3.5: Angular dependence of the standing-SAW-driven transition strengths as a function of the magnetic-field angle ϕ_B at a depth of 1000 nm inside SiC. Here, x_0 denotes the lateral position of the centre of the Gaussian laser spot relative to the chosen normal-strain antinode, and d_{FWHM} denotes the full width at half maximum of its intensity profile. The four panels show the V2 and V3 centres in their ground and excited states.

elaborate more on this, as we have two counterfacing IDTs, even if we power only a single IDT from this pair, the generated wave propagates towards the counterfacing IDT and is partially reflected back. The two IDTs form a cavity while focusing the SAW at the centre of the pair. A travelling-wave case would correspond to one propagation direction dominating over the counter-propagating wave, for example when one of the opposing IDTs generates a stronger SAW. Due to the aforementioned cavity effect, the signal amplitude is much higher in the standing-wave configuration, and, as this makes detection more reliable, we have decided to use this configuration for our experiments.

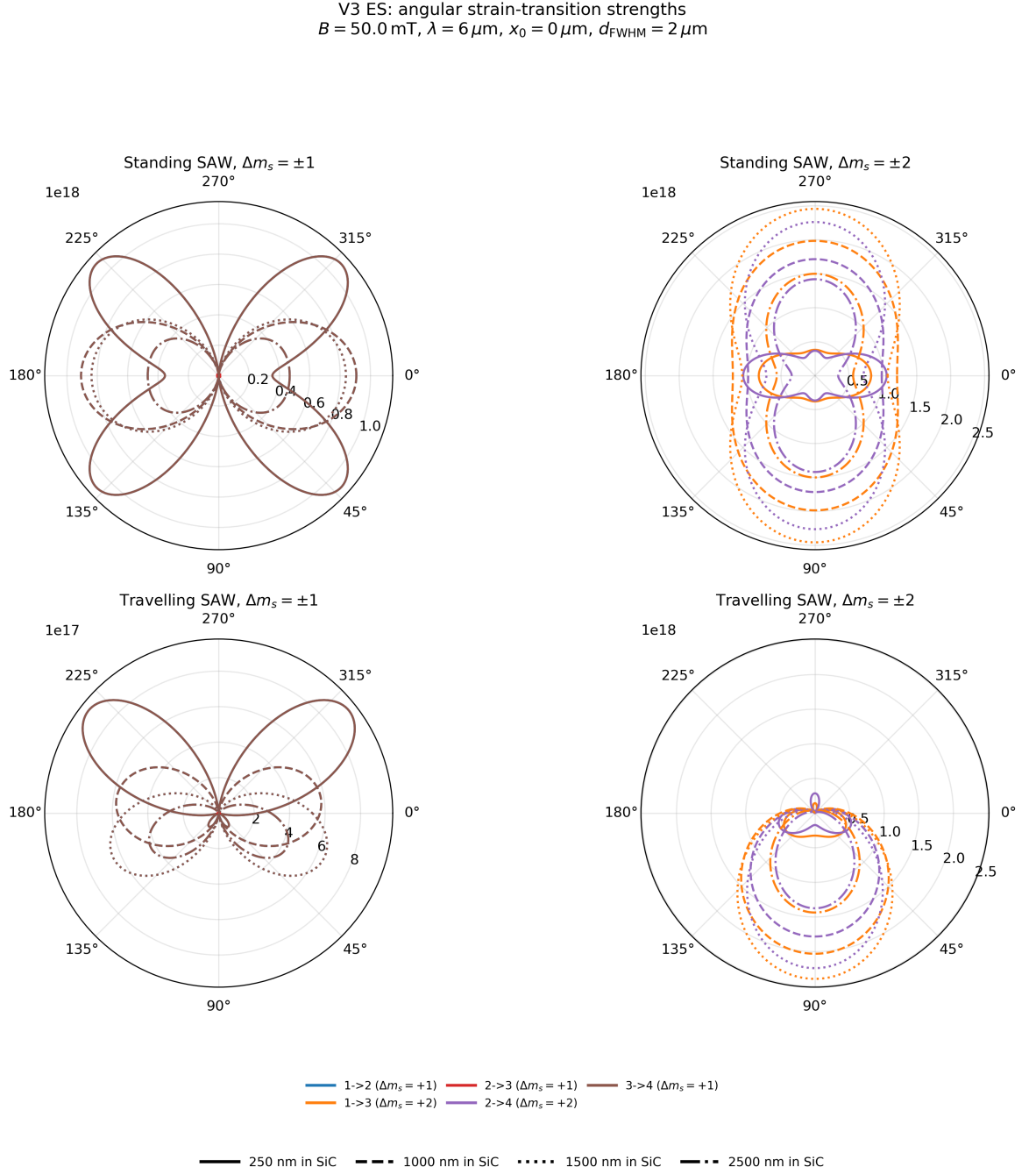


Figure 3.6: Angular dependence of the SAW-driven transition strengths for the V3 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $2 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

Chapter 4

Experimental Methods

4.1 6H-SiC and Colour Centres

As we have decided what depth our colour centres will be located we can start by working on our substrate. We have a 1cm x 1cm 6H-SiC with 500 μ m thickness from Wolfspeed, Inc branded as High Purity Semi Insulating (HPSI) SiC Substrate. Before starting on any process we shall measure and confirm the structural integrity of the substrate. We have two identical substrate which we process. This will give us flexibility in the laboratory in the case that one of the pair gets contaminated or damaged we can move on with the other without losing much time.

4.1.1 Raman Spectroscopy

One of the methods to confirm the structure of a substrate is to perform Raman Spectroscopy measurements. Raman spectroscopy is a technique which utilizes the phenomenon of Raman scattering. When a sample is irradiated with a laser beam of frequency ν_0 , majority of the scattered light is elastic, Rayleigh scattering, which has the same frequency as the incident light. A much weaker part of the scattered light is the so called Raman scattering. Instead of an elastic scattering, the frequency of the incident light is shifted to $\nu_0 - \nu_m$ or $\nu_0 + \nu_m$, where ν_m is the molecular vibrational frequency as shown in Figure 4.1. These shifts are called the Stokes and the anti-Stokes lines, respectively. Raman spectroscopy measures molecular vibrational frequencies as shifts from the incident laser frequency rather than direct absorption frequencies. As each different molecule or substrate has its own characteristic structure, it has characteristic vibrational modes, which appear at specific Raman shifts. This allows us to distinguish one substrate from another [48].

In order to explain the physical origin of Raman scattering, we can look at how the electric field of the laser interacts with a molecule. When the light reaches the molecule, its oscillating electric field induces an electric dipole moment in the molecule. The relationship between this induced dipole and the electric field is known as the polarizability of the molecule. If the molecule is vibrating, the nuclear displacement changes with the vibrational frequency, and because of this, the polarizability of the molecule can also change during the vibration [48].

This change in polarizability is very important, because it is the key requirement for a vibration to be Raman-active. In the classical explanation, one part of the induced dipole radiates light at the same frequency as the incident laser light, which gives Rayleigh scattering. Another part radiates light at shifted frequencies, $\nu_0 + \nu_m$ and $\nu_0 - \nu_m$, which gives anti-Stokes and Stokes Raman scattering. Therefore, if the polarizability does not change during the vibration, the vibration cannot be Raman-active [48].

In a Raman spectrum, the result is usually shown as intensity against Raman shift. The

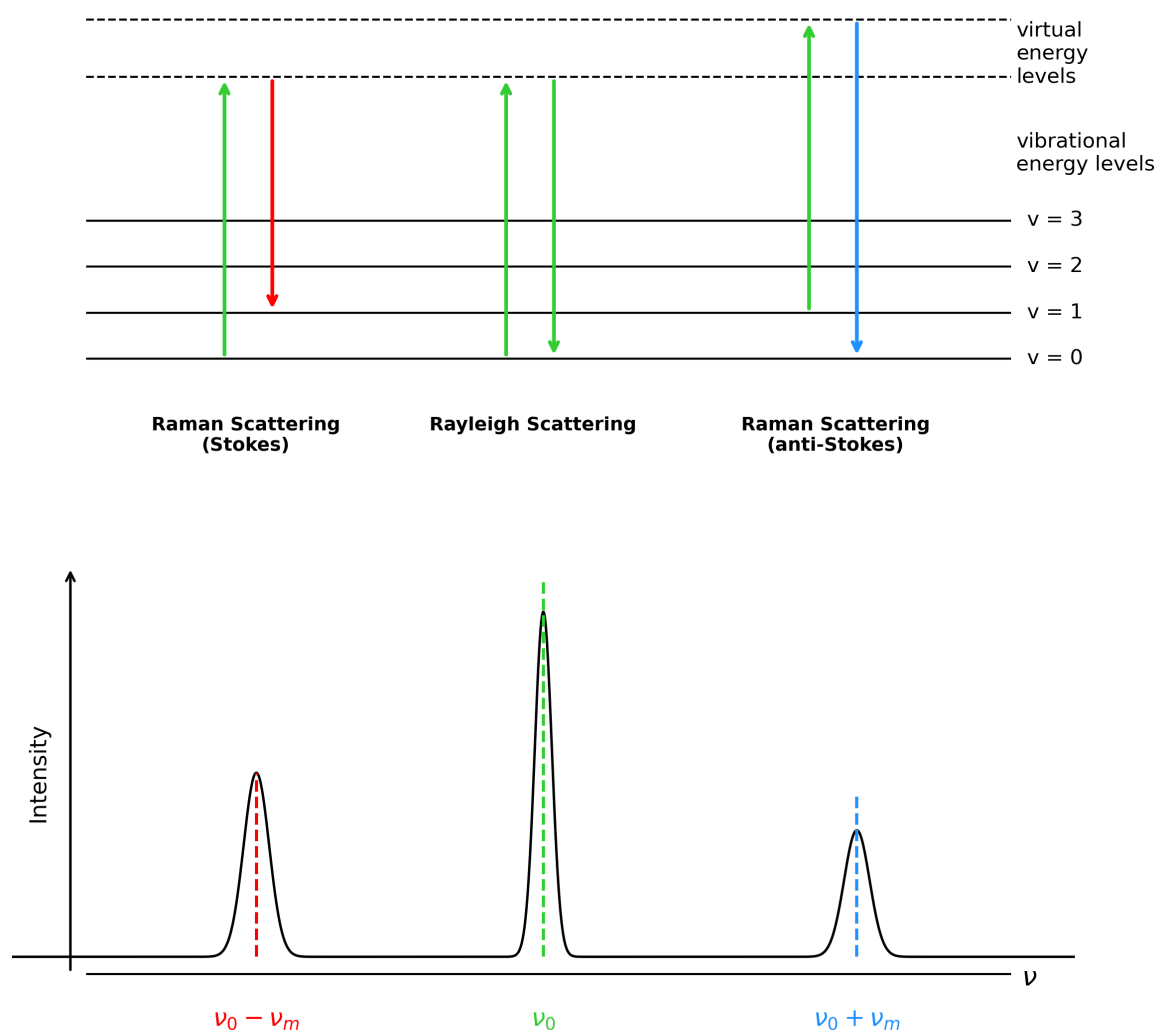
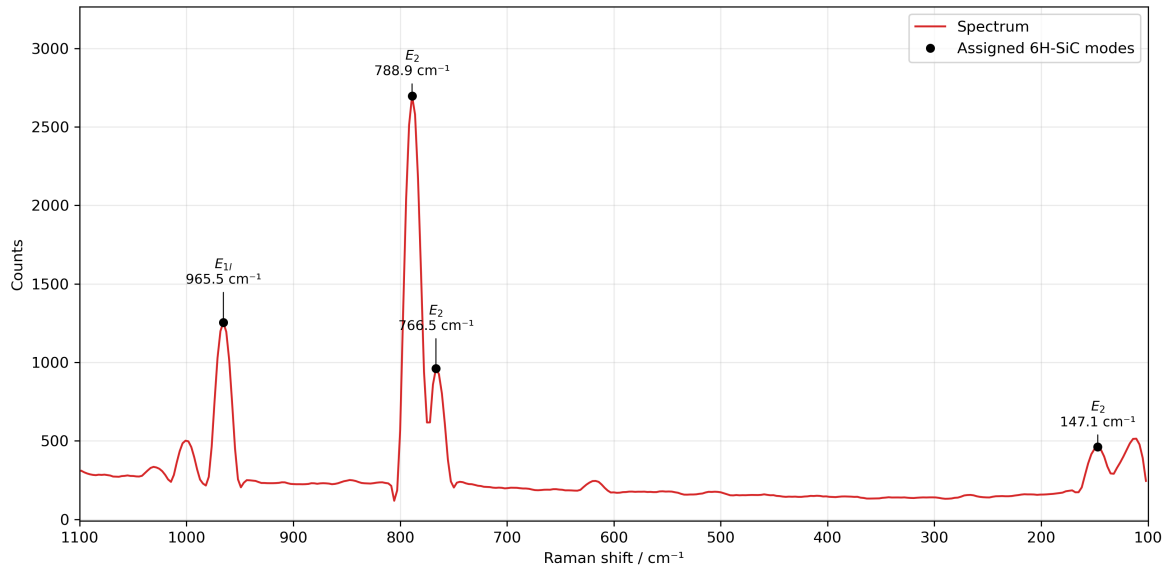


Figure 4.1: Schematic representation of Stokes (represented by the red line), Rayleigh (represented by the green line) and anti-Stokes (represented by the blue line) scattering. The incident laser radiation of frequency ν_0 interacts with the molecule and promotes it to a virtual energy level. In Rayleigh scattering, the scattered light has the same frequency as the incident light. In Stokes scattering, the scattered light has a lower frequency, $\nu_0 - \nu_m$, while in anti-Stokes scattering, the scattered light has a higher frequency, $\nu_0 + \nu_m$. The lower spectrum shows the corresponding intensity of the Rayleigh, Stokes, and anti-Stokes lines as a function of frequency.

Raman shift corresponds to the difference between the laser frequency and the frequency of the scattered light, and because of this, it directly represents the vibrational frequency of the

molecule. Under normal conditions, the Stokes lines are stronger than the anti-Stokes lines, because more molecules are found in the lower vibrational state than in the higher vibrational state. For this reason, Raman spectra are often measured on the Stokes side. Overall, this makes Raman spectroscopy very useful for identifying molecular vibrations and structural features [48].

With the assistance of Meysam Saeedi, we conducted a Raman scan for our 6H-SiC sample. In Figure 4.2a, we can see the result of our Raman spectrum scan and Figure 4.2b shows a reference Raman spectrum of 6H-SiC. The clear sharp peaks correspond to Raman-active vibrational modes of the investigated 6H-SiC sample, and their x-axis values indicate the Raman shifts in cm^{-1} . When we compare our measurements to the previous literature 4.2b, we can see that the E_{1l} mode and the three E_2 peaks shown in Figure 4.2b correspond well with the vibrational modes observed in our spectrum. Thus, the close agreement with the reference spectrum provides strong evidence that our sample is 6H-SiC.



(a) Raman spectrum of the investigated 6H-SiC sample.

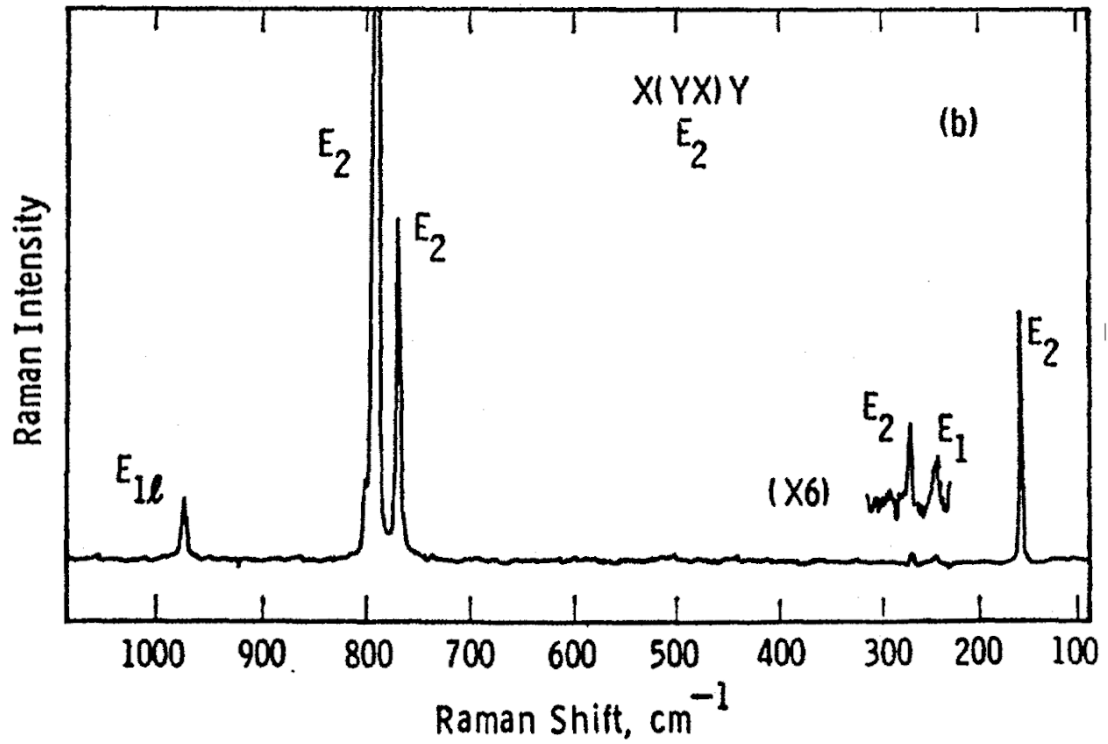
(b) Reference Raman spectrum of 6H-SiC showing the full range of the one-phonon emission region. The measurement geometry allows observation of the E_{1l} and E_2 modes [49].

Figure 4.2: Comparison of the measured and reference Raman spectra of 6H-SiC.

4.1.2 Creation of the V_{Si}

In Chapter 3.1, we have investigated the depth dependence of the SAW strain amplitude in 6H-SiC. Upon this investigation, we have decided on creating the colour centres at the depth of 1000 nm.

The V_{Si} centres will be created on the substrate via proton irradiation. In proton irradiation, energetic protons are implanted into the SiC substrate. In 6H- and 4H-SiC, room-temperature proton implantation has been shown to generate point defects which are attributed to silicon vacancies [50]. Furthermore, proton irradiation has been used for the controlled generation of optically active vacancy spins in SiC, where the generated centres were identified as silicon vacancies by their optical and magnetic resonance signatures [51]. As the protons travel through the crystal, they lose their energy through interactions with the electrons and atoms of the lattice. If enough energy is transferred to a lattice atom, the atom can be displaced from its original lattice site. This creates point defects such as vacancies and interstitials. In our case, the important defect is the silicon vacancy, where a silicon atom is removed from its lattice position, forming a V_{Si} centre. The implantation energy mainly determines how deep the protons penetrate into the substrate, while the fluence determines the number of implanted protons per unit area and therefore affects the amount of created defects.

Figure 4.3 shows the simulated V_{Si} as a function of depth after the proton irradiation. Simulation results showed us that 175 keV of applied energy results with the V_{Si} to be concentrated at our desired depth of 1000 nm. Furthermore, we chose a fluence of $1 \times 10^{15} \text{ cm}^{-2}$, since higher proton irradiation fluences reduce spin coherence and cause broadening that prevents the spin splittings from being resolved[52].

The sample has been sent to Dr. Andrei Anisimov at the Helmholtz-Zentrum Dresden-Rossendorf (HZDR) for the proton irradiation to be conducted.

The proton irradiation is conducted with the following parameters:

- Energy: 175 keV
- Fluence: $1 \times 10^{15} \text{ cm}^{-2}$
- Temperature: Room temperature
- Side for implementation: Si-side

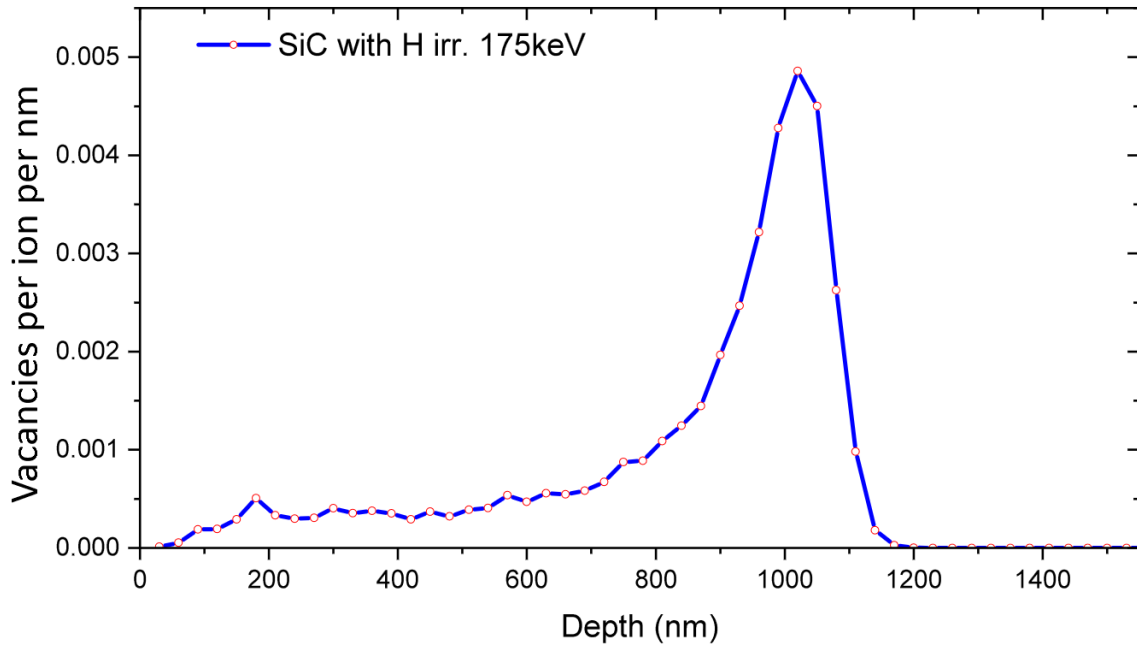


Figure 4.3: Simulated Si-vacancy distribution as a function of depth after H ion implantation in 6H-SiC with an implantation energy of 175 keV. The simulation was provided by Dr. Andrei Anisimov.

4.1.3 Photoluminescence Scan

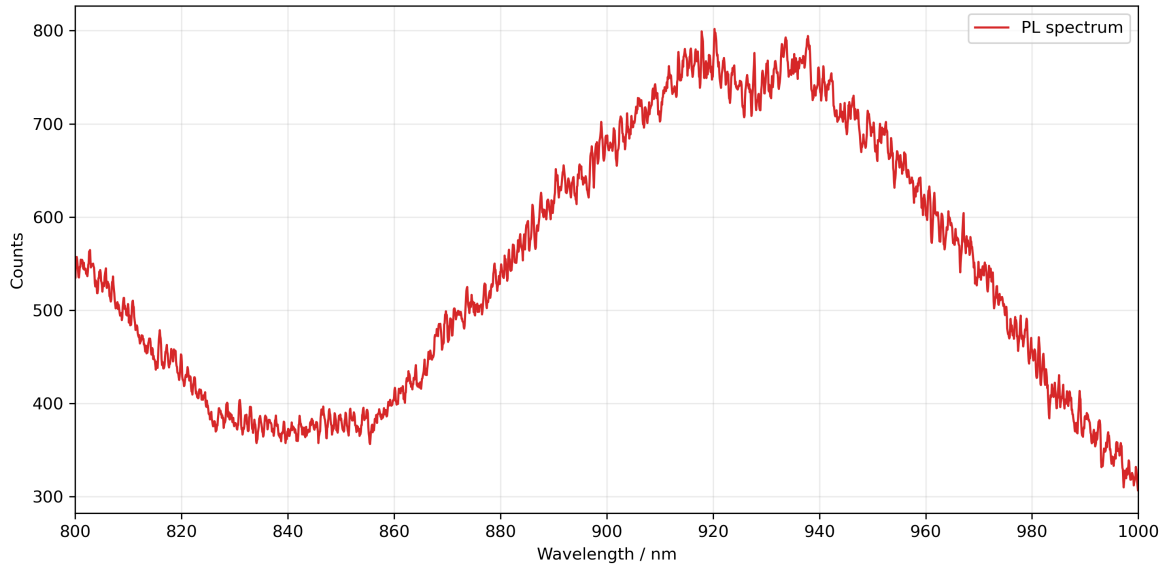
Furthermore we can investigate the substrate in terms of its photoluminescence properties. One of the methods to investigate optically active defects in a substrate is to perform photoluminescence spectroscopy measurements. Photoluminescence spectroscopy, which is usually abbreviated as PL spectroscopy, is a technique in which the sample is excited by light and the light emitted from the sample is measured. When the sample absorbs photons from the excitation laser, electrons can be promoted to higher energy states. After this excitation, the system relaxes back to a lower energy state, and part of this relaxation can happen by emitting photons. These emitted photons form the photoluminescence signal [53].

In other words we can say that, while Raman spectroscopy gives information about vibrational modes through Raman shifts, photoluminescence spectroscopy gives information about optical transitions and defect-related emission in the material. In semiconductors such as SiC, the emitted light can originate from radiative recombination processes or from optically active defect states inside the band gap. Therefore, PL spectroscopy is especially useful when we want to investigate color centers or defect-related optical emission in the substrate [53, 12].

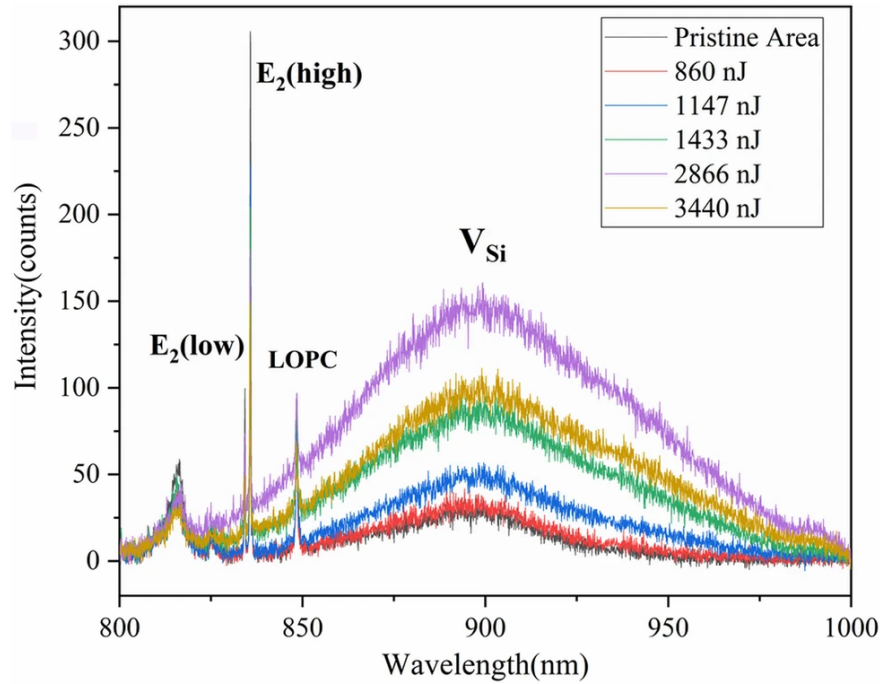
For our 6H-SiC sample, the photoluminescence scan is important because silicon vacancy

color centers in 6H-SiC can be detected through their PL signal. In the literature, Zhou et al. used room temperature photoluminescence measurements to characterize femtosecond-laser-written silicon vacancy color centers in 6H-SiC, and the PL signal was used to analyze the formation of the V_{Si} defects [12]. Therefore, by comparing our PL scan with previously confirmed literature, we can investigate whether the expected defect-related emission is present in our sample.

In Figure 4.4a, we can see the photoluminescence intensity as a function of wavelength at room temperature. We can compare this measurement with the measurement of Zhou et al. shown in Figure 4.4b. Here, one can observe a similar spectral shape between 800 nm and 1000 nm, which supports our previous intuition that the substrate contains 6H-SiC V_{Si} centres.



(a) Photoluminescence spectrum of the investigated 6H-SiC sample showing emission features related to V_{Si} colour centres, measured at room temperature using a 532 nm laser.



(b) Reference photoluminescence spectrum of V_{Si} centres in 6H-SiC at room temperature. The broad emission around 900 nm is associated with the silicon-vacancy signal, while the different curves show the change in photoluminescence intensity for different writing-pulse energies [12].

Figure 4.4: Comparison of the measured and reference photoluminescence spectra of silicon-vacancy colour centres in 6H-SiC.

4.1.4 Photoluminescence Spectra at Cryogenic Temperatures

Figure 4.4a shows the PL spectrum of 6H-SiC measured at room temperature. At room temperature, a broad emission band centred at approximately 900 nm is observed. As the temperature is reduced, zero phonon line (ZPL) and phonon sideband (PSB) becomes more clearly resolved.

For the spatially resolved PL measurements, the excitation beam was positioned along the x -, y -, and z -axes. The x - and y -positions were fixed at the centre between two IDTs, while the microscope-stage position along the z -axis was varied. Measurements were performed at nominal stage positions of $z = 10 \mu\text{m}$ and $z = 70 \mu\text{m}$, as shown in Figures 4.6 and 4.5, respectively.

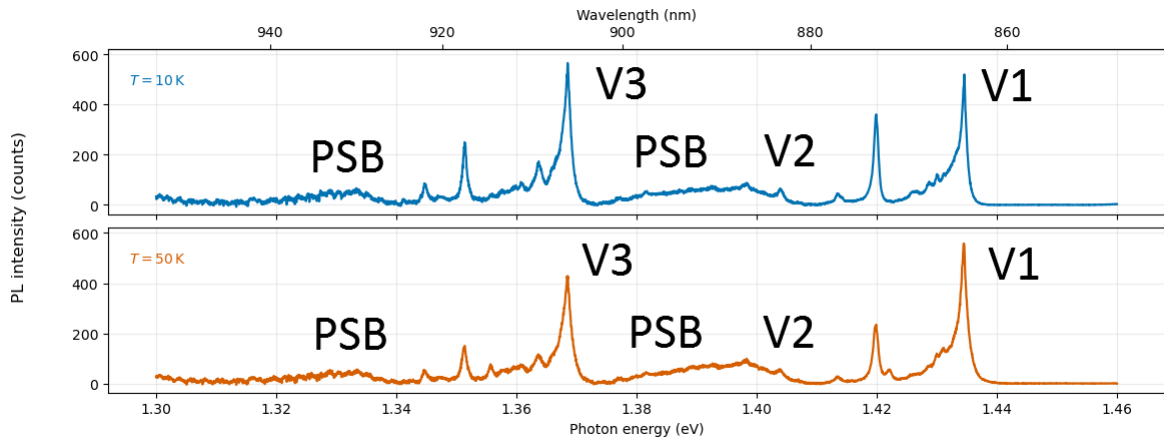


Figure 4.5: Temperature-dependent PL spectra of 6H-SiC measured at a depth of $z = 70 \mu\text{m}$, shown as a function of photon energy and wavelength.

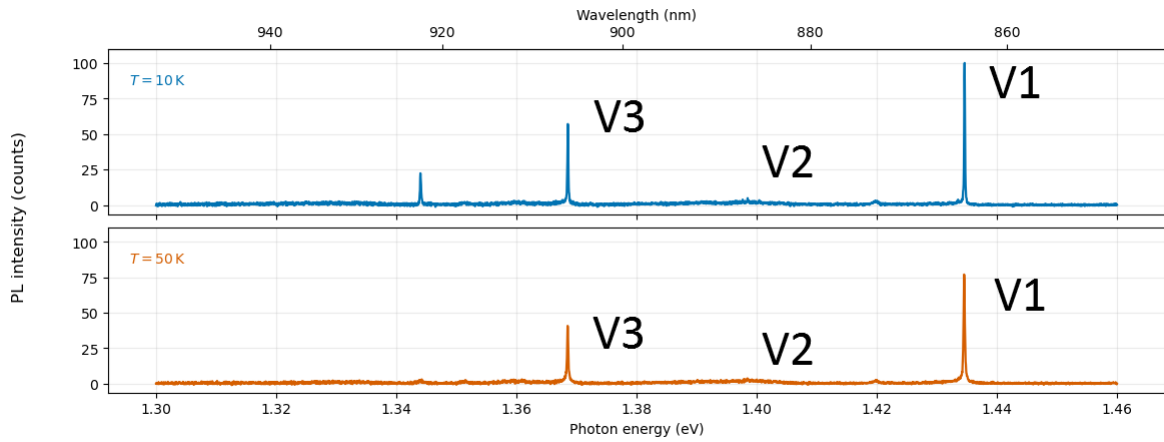


Figure 4.6: Temperature-dependent PL spectra of 6H-SiC measured at a depth of $z = 10 \mu\text{m}$, shown as a function of photon energy and wavelength.

A clear difference in both PL intensity and spectral shape is observed between the two z -positions. The PL spectra at $z = 70 \mu\text{m}$ is for the laser beam focused at the sample depth where a large density of V_{Si} were created via proton irradiation. The PL spectra at $z = 10 \mu\text{m}$ is with the laser focused deeper into the SiC and the PL is that of the low density of V_{Si} present naturally in SiC. Therefore, changing the z -position affects both the detected PL intensity and the relative visibility of the spectral features.

The narrow peaks observed at $z = 10 \mu\text{m}$ occur at 1.435, 1.398, and 1.369 eV. These energies agree closely with the zero-phonon lines reported by Wagner et al. [3] for the silicon-vacancy centres in 6H-SiC, which is represented on the Figure 4.7. Based on this spectral agreement, the observed peaks are assigned as follows:

- 1.435 eV: $V1/V1'$,
- 1.398 eV: $V2$,
- 1.369 eV: $V3$.

The measurements were conducted between 50 K and 10 K. At lower temperatures, the suppression of thermal broadening allows the zero-phonon lines to be resolved more clearly.

Breev et al. report that the polarization responses of the $V1$ and $V3$ centres cannot be resolved individually at room temperature. Their combined contribution to $\Delta\text{PL}/\text{PL}$ is predominantly polarized perpendicular to the crystal c -axis ($e_x, e_y \perp c$). At $T = 15 \text{ K}$, however, the $V1$, $V2$, and $V3$ silicon-vacancy (V_{Si}) centres in 6H-SiC can be spectroscopically distinguished through their respective zero-phonon lines [1].

In Figure 4.6, the $V1/V1'$, $V2$, and $V3$ emission lines can be identified, although their intensities differ considerably. In particular, the $V2$ line is substantially weaker than the $V1/V1'$ and $V3$ lines. As discussed in Chapter 2.1.3, this difference is consistent with the polarization-dependent emission properties reported by Breev et al. [1]. The $V2$ centre emits preferentially in the sample plane, whereas the present setup primarily detects the out-of-plane emission. In contrast, the emission patterns of the $V1/V1'$ and $V3$ centres provide a stronger contribution in the detection direction. This polarization dependence is therefore consistent with the comparatively weak $V2$ peak observed in the measured PL spectra.

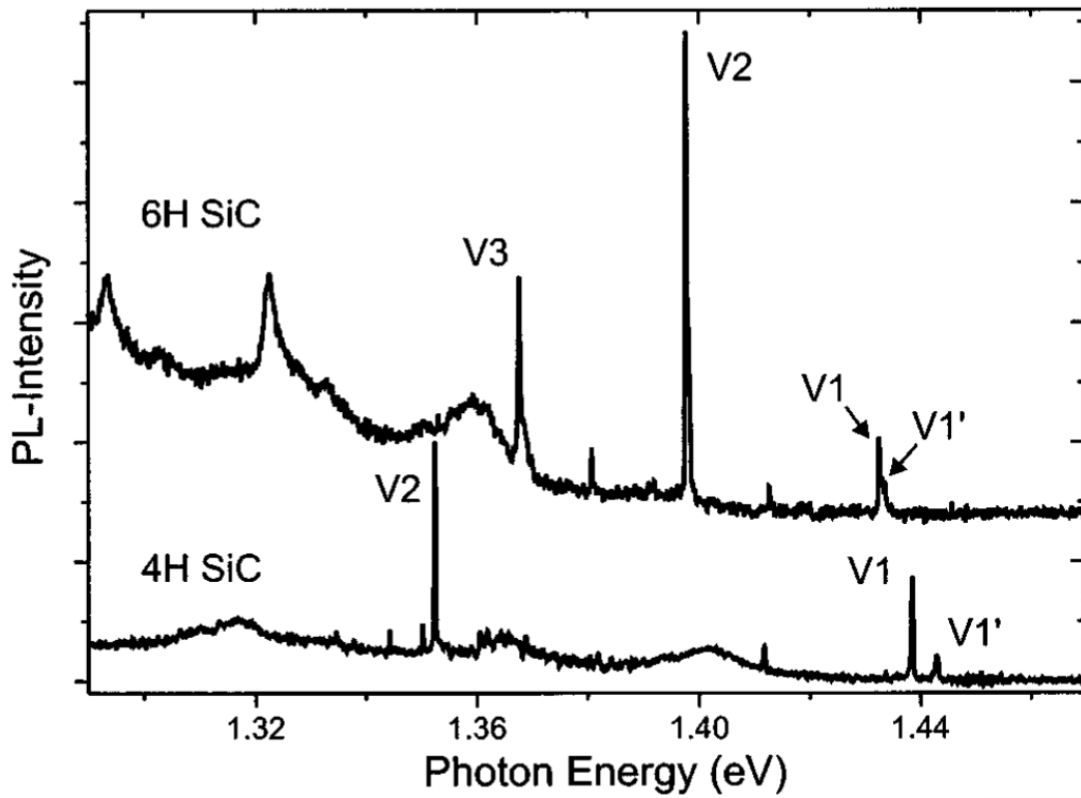


Figure 4.7: Photoluminescence spectra of electron-irradiated 6H-SiC (upper spectrum, $T = 10$ K) and 4H-SiC (lower spectrum, $T = 34$ K). Three no-phonon lines (V1, V2, and V3) are observed in 6H-SiC, whereas two (V1 and V2) are observed in 4H-SiC. The phonon-assisted sidebands appear at lower photon energies. Adapted from Wagner et al. [3].

4.2 Nanofabrication

With the V_{Si} centres created, we can continue with the nanofabrication processes. "Because SiC does not possess strong piezoelectric properties, we cannot achieve efficient piezoelectrically excited SAWs using SiC alone [54]. For the SAWs to be efficiently piezoelectrically excited, we need to create a suitable medium. An effective approach involves utilizing a layered SAW configuration, where an AlN or ZnO piezoelectric thin film is integrated onto a high-acoustic-velocity substrate [55]. ZnO is one of the suitable candidates. Due to its crystal structure, ZnO lacks a centre of symmetry, resulting in internal electric dipoles which shifts when the material is under mechanical stress [56]. ZnO thin films are especially useful for this purpose because they have good piezoelectric properties and a high electromechanical coupling coefficient. Furthermore, ZnO can be deposited as a thin film on different substrates, including silicon, which makes it suitable for integration with microfabrication processes and control electronic circuitry. Other advantages of ZnO are its good bonding with different substrate materials, high temperature stability, and relatively low deposition cost [37, 36]. Therefore, ZnO is a suitable choice for our device structure. We will create the ZnO layer using sputtering techniques.

On top of this, we shall place IDTs in order to generate SAWs, as explained in Section 2.3.2. This will be conducted using laser lithography techniques.

The steps toward establishing the experimental setup consist of the following processes:

- Sputtering
 - SiO_2 : 35 nm
 - ZnO : 700 nm
- Laser lithography
 - Patterning of the IDT mask
- Metal evaporation

4.2.1 Sputtering

Sputtering is a thin film deposition technique which belongs to the physical vapour deposition methods. In this technique, the sample is placed inside a vacuum chamber together with the target material which we want to deposit. After introducing a process gas, usually argon, a plasma is created inside the chamber. The positive ions from the plasma are accelerated

towards the target and hit its surface. Due to this bombardment, atoms from the target are ejected and travel through the chamber. These atoms then condense on the substrate surface and form a thin film. In this way, materials such as SiO_2 and ZnO can be deposited with controlled thickness on the sample [57]. SiO_2 is used as a thin intermediate layer to improve the adhesion of ZnO to the substrate surface.

We took our sample to the clean room and placed it into the sputtering chamber. We have two identical samples of 6H-SiC which can be simply named as sample A and sample B. The temperature was lowered for the sputtering of Sample B. We conducted the following recipe for the sputtering:

- Target: 35 nm SiO_2 + 700 nm ZnO
- Temperature (T):
 - Sample A: 350°C for SiO_2 and ZnO
 - Sample B: 200°C for SiO_2 and ZnO
- $z = 65$ mm for SiO_2 and ZnO
- $P = 80$ W for SiO_2 and ZnO
- $t = SiO_2 \rightarrow 2562$ s, $ZnO \rightarrow 12787$ s
- $p =$ Butterfly on 11% for SiO_2 and ZnO

Here, the target defines the intended deposited layer thicknesses, namely 35 nm of SiO_2 and 700 nm of ZnO . The parameter T refers to the chamber temperature during sputtering. The parameter z refers to the distance between the sputtering target and the sample, which was set to 65 mm. The value 80 W refers to the sputtering power used for both SiO_2 and ZnO deposition. The parameter t refers to the deposition time, which was 2562 s for SiO_2 and 12787 s for ZnO . Finally, the parameter p refers to the butterfly valve setting of the sputtering chamber. The butterfly valve controls the pumping speed and therefore helps to regulate the chamber pressure during deposition. For both SiO_2 and ZnO , the butterfly valve was kept at 11% open.

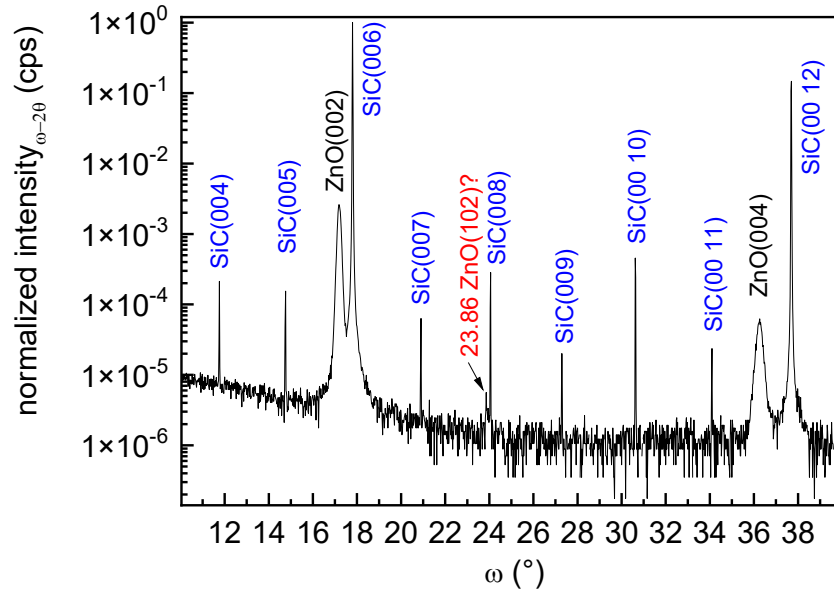
4.2.2 X-Ray Scan of the Substrate

In order to confirm the precision of our sputtering process, we have conducted an X-ray diffraction measurement of our substrate. X-ray diffraction, usually abbreviated as XRD, is a method used to investigate the crystalline structure of a material. In this technique, X-rays are directed onto the sample, where they interact with the crystal planes of the material. If the condition for constructive interference is fulfilled, diffraction peaks appear at specific angles. This condition is described by Bragg's law, and the peak positions can therefore be related to the spacing between crystal planes. In this way, XRD allows us to identify the crystal structure, orientation, and quality of the deposited ZnO layer [58]. This is important after sputtering because a highly *c*-axis-oriented ZnO layer, perpendicular to the sample surface, is essential to maximize the electromechanical coupling and overall piezoelectric response [37].

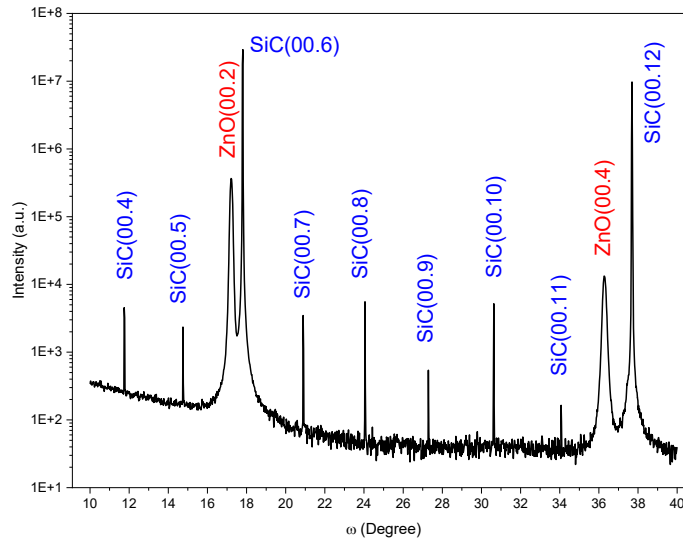
Fig. 4.8a shows the X-ray diffraction (XRD) ω - 2θ scan of the 6H-SiC substrate after ZnO thin-film deposition. In an ω - 2θ scan, both the sample and the detector are rotated during the measurement. The sample is rotated by the angle ω , while the detector is moved by the angle 2θ . This means that the detector angle is kept twice as large as the sample angle. This geometry is used so that the diffracted X-rays can be detected at the correct diffraction angle while different lattice spacings are scanned. Therefore, the angles where strong diffraction peaks appear give information about the lattice spacing and the crystallographic planes present in the sample.

The pattern, plotted on a logarithmic intensity scale, exhibits dominant (00 l) reflections of 6H-SiC (004), (005), (006), (008), (0010), (0011), and (0012), confirming the strong *c*-axis orientation and high crystalline quality of the substrate. Distinct ZnO (002) and (004) reflections are clearly observed, consistent with the intended growth of a *c*-axis-oriented ZnO layer on the SiC substrate. The sharp and well-defined diffraction peaks demonstrate good crystallinity, while slight peak shifts suggest the presence of strain in the ZnO film. Comparison with previous reports [59, 60] enables identification of the observed crystallographic planes and their corresponding Miller indices.

For further assessment of the crystalline quality of the ZnO layer, an ω -scan, also called a rocking curve measurement, was performed. In this measurement, the diffraction angle is kept fixed around the selected ZnO (002) reflection, while the sample is slightly rotated around the ω angle. Therefore, the ω -scan does not mainly identify new phases, but instead shows how well the selected crystal planes are aligned. A narrow rocking curve indicates good out-of-plane alignment, while a broader curve indicates crystallite tilt and mosaic spread. On the literature, high *c*-axis textured films with rocking curve full width at half maximum (FWHM) of 1.69° was demonstrated [61]. As shown in Fig. 4.9a, the relatively large ω -scan



(a) XRD ω - 2θ scan of the 6H-SiC substrate with a ZnO thin film, plotted on a logarithmic intensity scale. The notation (006) denotes the measured crystallographic reflection of 6H-SiC. Measurement performed by Dr. Klaus Biermann.



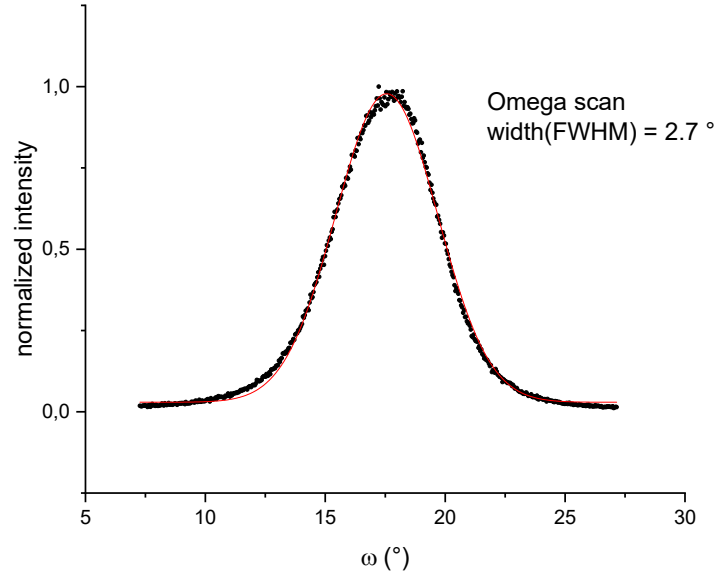
(b) XRD ω - 2θ scan of a 6H-SiC substrate with a ZnO thin film deposited at reduced substrate temperature. Measurement performed by Dr. Michael Hanke.

Figure 4.8: Comparison of XRD ω - 2θ scans of ZnO thin films deposited on 6H-SiC substrates under different conditions.

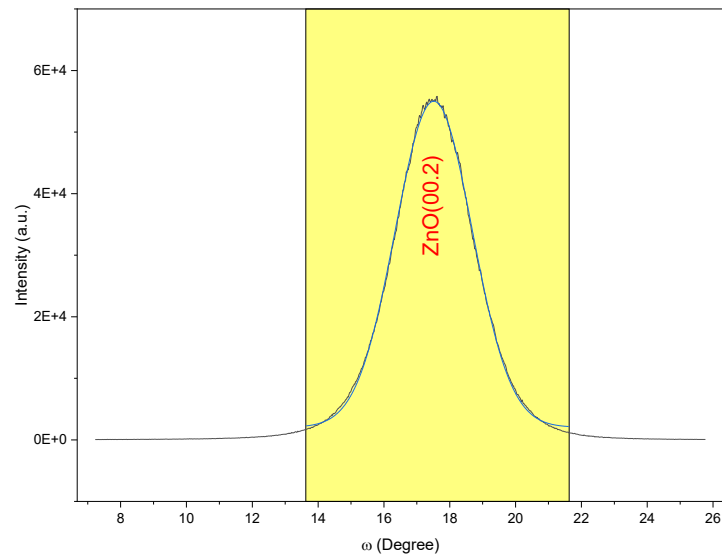
width suggests a considerable mosaic spread in the ZnO layer. The extracted FWHM of 5.02° therefore indicates a degree of non-ideal crystalline alignment. Since smaller rocking-curve FWHM values correspond to better crystalline alignment, this parameter is useful for comparing the crystalline quality of different ZnO layers [56]. Although the rocking curve is relatively broad, the ZnO crystallites are still expected to have a preferred c-axis orientation approximately perpendicular to the substrate surface. Therefore, for the first substrate, we proceeded with the fabrication by applying the IDT mask on top of the ZnO layer and experimentally evaluating the device performance.

At a later stage, a second sputtering process was conducted on sample B with a reduced substrate temperature of 200°C . This was done in order to investigate whether lowering the sputtering temperature could improve the crystalline quality of the ZnO layer. For the first substrate, which was sputtered at a substrate temperature of 350°C , the ω -scan of the ZnO layer gave an FWHM of 5.02° . For the second substrate sputtered at 200°C , we obtained a much better result with an FWHM of approximately 2.7° , as can be seen in Fig. 4.9b. Based on the reduced FWHM of the ZnO (002) reflection, the layer quality seems to have improved compared to the previous measurement.

Also, in Figure 4.8b, we can see that the small peak which was suspected to result from the ZnO (102) orientation in Figure 4.8a is no longer apparent. This further supports that the reduced sputtering temperature improved the crystalline quality of the ZnO layer.



(a) ω -scan of the ZnO (002) peak with Gaussian fit, yielding a FWHM of 5.02° , indicating noticeable mosaic spread. Measurement performed by Dr. Klaus Biermann.



(b) ω -scan of the ZnO (002) peak with Gaussian fit, yielding a FWHM of approximately 2.7° . Measurement performed by Dr. Michael Hanke.

Figure 4.9: Comparison of ω -scan rocking curves of the ZnO (002) peak: (a) FWHM of 5.02° and (b) FWHM of approximately 2.7° .

4.2.3 Design of the Interdigital Transducers (IDTs)

In our design, we employ two sets of SAW delay lines oriented at 90° to each other, with each delay line comprising two opposing focusing IDTs, to generate linearly independent strain fields at the focal point.

A SAW propagating along x predominantly generates $\{\epsilon_{xx}, \epsilon_{xz}, \epsilon_{zz}\}$, while a SAW propagating along y generates $\{\epsilon_{yy}, \epsilon_{yz}, \epsilon_{zz}\}$. Because these strain components couple to different combinations of the spin operator components, the two orthogonal IDTs provide independent control over out-of-plane (S_z^2) and in-plane spin-mixing terms¹. This adds an additional degree of freedom to the spin-driving protocol and therefore extends our experimental freedom. Experimentally, we mostly use a single pair of IDTs placed along the same axis. The

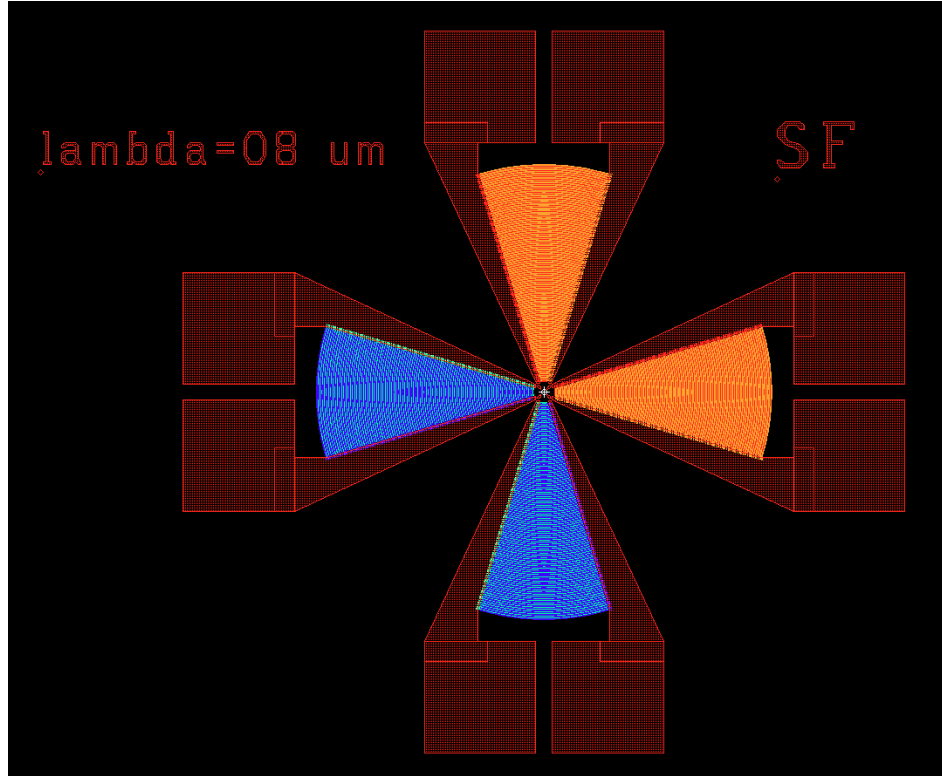


Figure 4.10: Schematic of the four-port interdigitated transducer (IDT) structure designed to generate orthogonal surface acoustic waves (SAWs) with the acoustic wavelength $\lambda = 8\mu m$, corresponding to the designed operating frequency.

additional IDT pairs give us more flexibility within the confined device area and provide backup structures in case some IDTs get damaged.

¹In our experiments, the longitudinal S_z^2 term is not directly exploited, since the magnetic field is applied in the plane. However, this property of the structure could still be useful for other experimental configurations.

On Fig. 4.10 we show the structure of the device. Each interdigitated transducer (IDT) consists of 200 single-finger electrode configuration designed to generate surface acoustic waves (SAWs). For the $4\mu\text{m}$, $6\mu\text{m}$, and $8\mu\text{m}$ IDTs, the distances from the closest point of the final finger to the focal point are $20\mu\text{m}$, $24\mu\text{m}$, and $32\mu\text{m}$, respectively. The corresponding aperture widths are $16\mu\text{m}$, $24\mu\text{m}$, and $32\mu\text{m}$, respectively.

The fingers are connected to individual feed lines, which terminate in square contact pads. These contact pads serve as connection points for the incoming RF signals.

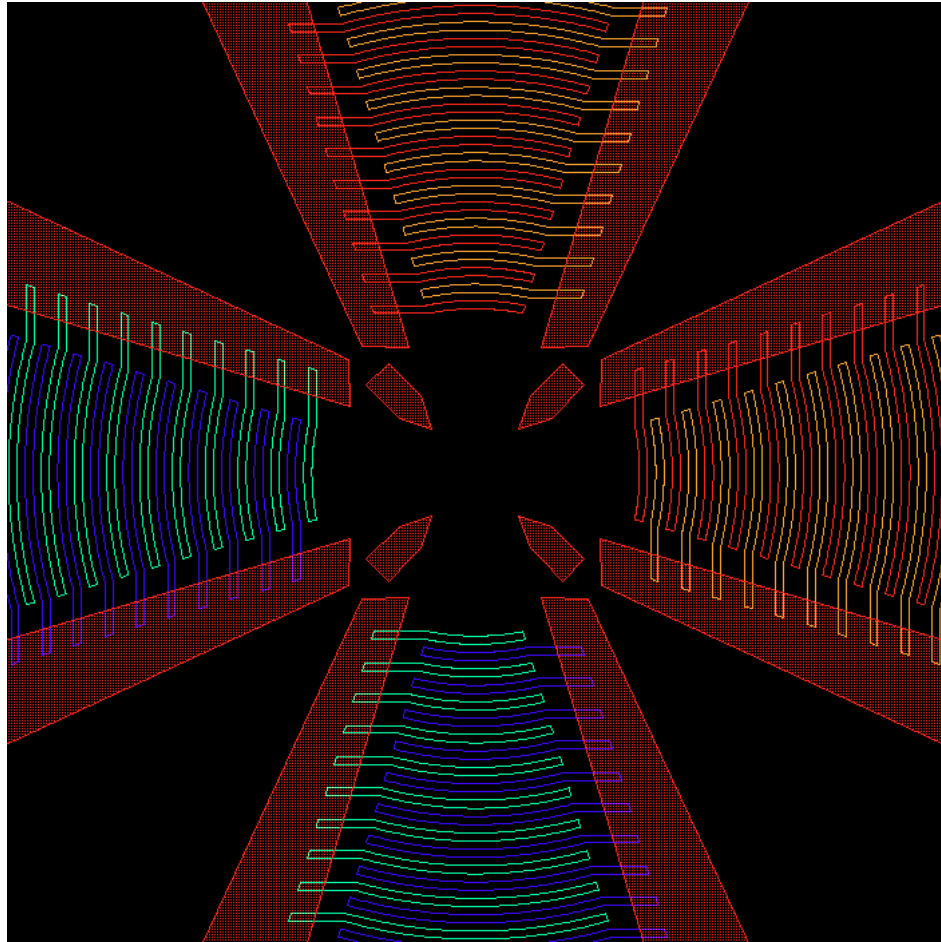


Figure 4.11: Zoomed view of the focal region of the focused interdigitated transducers (IDTs).

On Fig. 4.11 we can see the focal point of our IDT. The wavelength is determined by simply adjusting the gap between the individual fingers. This system uses single finger (SF) fringe system for wave generation. One may also notice the arrow-like structures pointing towards the centre. The arrow-shaped structures pointing toward the centre serve as visual alignment markers for precise laser positioning during measurements. The mask contains two orthogonal delay lines, each comprising three acoustic wavelengths: $4\mu\text{m}$, $6\mu\text{m}$, and

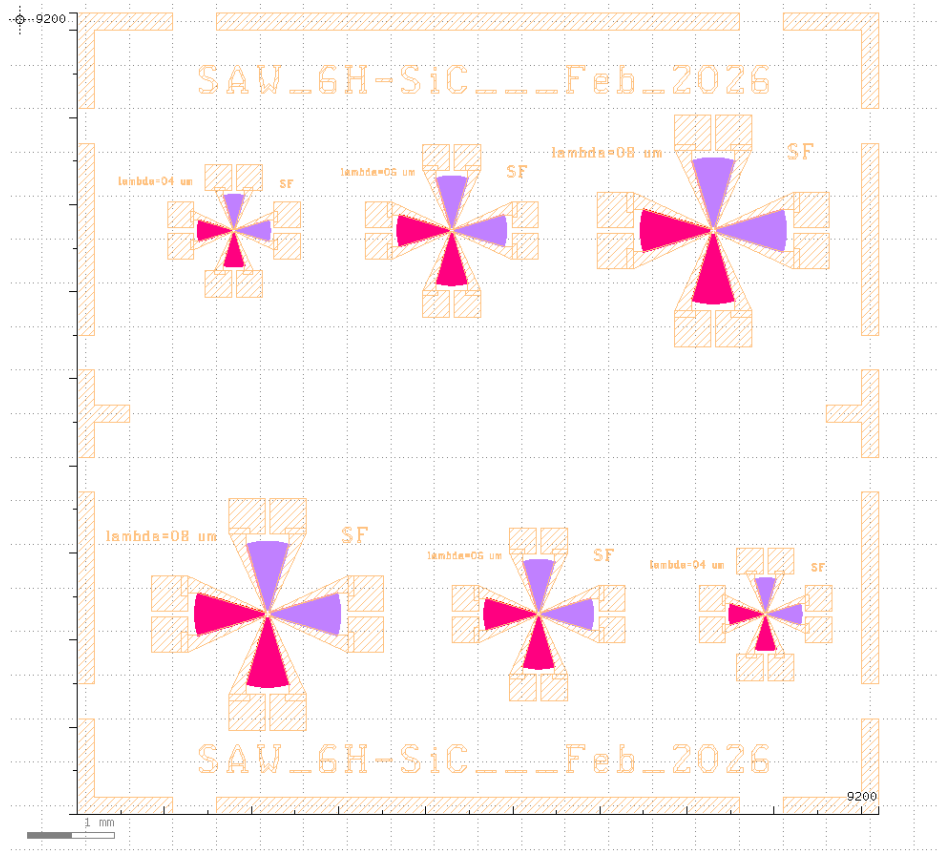


Figure 4.12: Mask layout of the 6H-SiC surface acoustic wave (SAW) device, showing multiple focused interdigitated transducer (IDT) designs with wavelengths $\lambda = 4 \mu\text{m}$, $6 \mu\text{m}$, $8 \mu\text{m}$ arranged for fabrication on a single chip.

$8 \mu\text{m}$. Fig. 4.12 shows the complete mask layout. The square pads surrounding the IDTs serve as contact pads for RF connections. The spacing between adjacent contact pads is fixed at $60 \mu\text{m}$. As a consequence, the connection pad geometry appears disproportionate for the different wavelengths. A clear gap is introduced along the horizontal axis through the centre of the mask to facilitate cutting of the substrate, allowing the fabrication of two samples from a single $10 \times 10 \text{ mm}^2$ piece.

4.3 Laser Lithography and Metal Evaporation

Laser lithography is a direct writing technique used to transfer a designed structure onto the sample surface. In this method, the sample is coated with a photoresist layer, and the desired pattern is written by scanning a focused laser beam over the substrate. Dresel and Schwider describe this principle using a commercial laser lithography system, where the laser beam

is modulated and scanned over the substrate in order to expose the desired pattern [62]. After the exposure, the photoresist is developed, so that the written pattern is opened on the sample surface. In our case, this process is used to define the IDT geometry before the metal deposition step.

After the lithography step, metal evaporation is used to deposit the conducting electrode material. Metal evaporation is a physical vapour deposition technique in which the metal source is heated inside a high-vacuum chamber until metal atoms evaporate from the source. These atoms travel through the chamber and condense on the sample surface, forming a thin metal film. Since the metal mainly deposits in a line-of-sight direction, this method is suitable for transferring the lithographically defined IDT pattern onto the substrate [57]. In our case, a Ti/Al/Ti layer is deposited where the first Ti layer is an adhesion layer whilst the second Ti layer prevents Al oxidation. The Al layer forms the main conducting part of the IDTs. Ti/Al-based metallization stacks are also commonly used for electrical contacts on SiC [63].

Before the lithography process, the sample surface was cleaned with acetone and isopropanol in order to remove organic residues, dust particles, and other surface contamination. This cleaning step is important because contamination can prevent the uniform coating of the adhesion promoter and the photoresist. Afterwards, the adhesion promoter was applied in order to improve the attachment of the photoresist to the sample surface. The photoresist was then spin-coated, and the sample was heated on a hot plate to remove solvent from the resist and to stabilize the resist layer before exposure. After the laser writing, the sample was heated again as a post-exposure bake in order to support the development of the exposed photoresist pattern.

After this, the exposed pattern was developed using AZ 726 MIF developer. The development step removes the exposed regions of the photoresist and opens the IDT pattern on the sample surface. Before metal evaporation, oxygen plasma cleaning was used to remove remaining resist residues from the opened areas, so that the metal could contact the surface more cleanly. After the Ti/Al/Ti evaporation, the final acetone bath and ultrasonic cleaning were used for the lift-off process. In this step, the remaining photoresist and the unwanted metal on top of it are removed, leaving only the metal IDT structure on the sample.

We took our sample to the clean room and conducted the following recipe for the laser lithography and metal evaporation:

- Acetone bath - 3-5 min
- Ultrasonic bath in acetone - 2 min
- Isopropanol rinse - 30-60 s
- Drying with nitrogen

- AR300-80 Adhesion Promoter: ~ 1 nm
 - 4000 RPM - 40 s
 - Heating on hot plate: 150°C - 3 min
 - Acetone bath - 3-5 min
 - Isopropanol rinse - 30-60 s
- AZ MiR 701 photoresist diluted with AZ EBR solvent at a volume ratio of 3 : 2:
 ~ 300 nm
 - 4000 RPM - 40 s
 - Heating on hot plate: 90°C - 1 min
- Laser writer - *Heidelberg DWL66+*
 - Settings:
 - * Optical auto focus: 40%
 - * Focus offset: 10%
 - * Intensity: 65%
 - * Filter: 12.5%
 - * Laser power: 20 mW
 - Heating on hot plate: 110°C - 1 min
- AZ 726 MIF developer diluted with deionised water at a volume ratio of 4 : 1 – 40 s
- De-ionised water bath – 120 s
- Plasma asher, using oxygen plasma to clean the surface
- Metal evaporation - *Pfeiffer Vacuum Classic 500*: $\sim 10^{-6}$ mbar
 - Ti: 10 nm
 - Al: 30 nm
 - Ti: 10 nm
- Final cleaning
 - Acetone bath – 8 min
 - Ultrasonic bath – 1 min

- Isopropanol rinse – 30–60 s

In Figure 4.13, we can see our sample just before the metal evaporation process. As it can be seen more clearly in Figure 4.14, there was some residue near the IDT fingers. At this point, we were not sure whether this was surface contamination or whether the contamination had occurred during or before the sputtering process. Therefore, we cleaned the surface of the substrate again with acetone and isopropanol and re-conducted the entire process. After that, although the sample was not entirely free from contamination, we were satisfied with the result and decided to continue with the next steps.

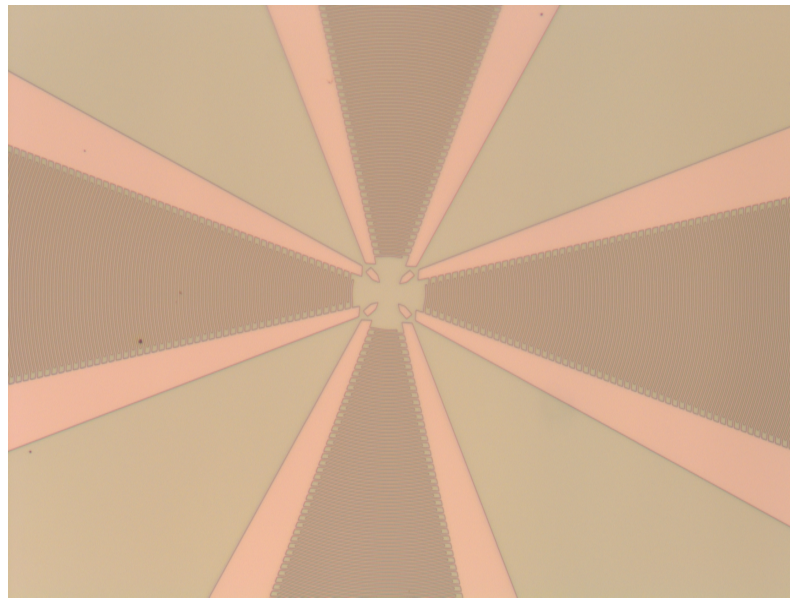


Figure 4.13: Sample after photoresist coating and laser writing, just before the metal evaporation process.



Figure 4.14: Residue or defect observed near the edge of the IDT fingers before metal evaporation.

Figure 4.15 and the following figures show the sample after the metal evaporation process. In Figure 4.15, the “L” shaped scratch on the left corner of the sample is used as a sign to indicate the top surface of the substrate. Since it is located on the back side of the sample, it does not appear as a defect on the actual device structure. The colour shifting pattern on the top right corner is the region where the holding arm rested during the ZnO sputtering process. Although this deformation is on the front face of the sample, it does not overlap with the IDTs and therefore does not harm the functionality of the sample.

In Figure 4.17, we can see small spots near the beginning and end of some individual fingers. Nevertheless, in the frequency response tests presented in the next chapter, we observe no overall dysfunctionality of the IDTs. All of them respond in the expected frequency ranges.

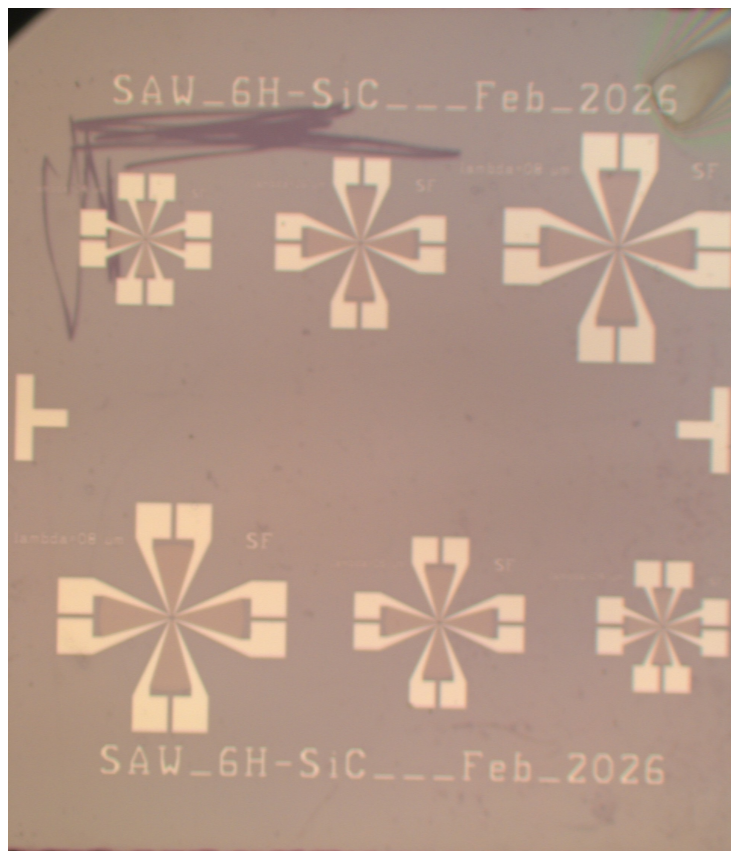


Figure 4.15: Sample after metal evaporation, inspected under the microscope.

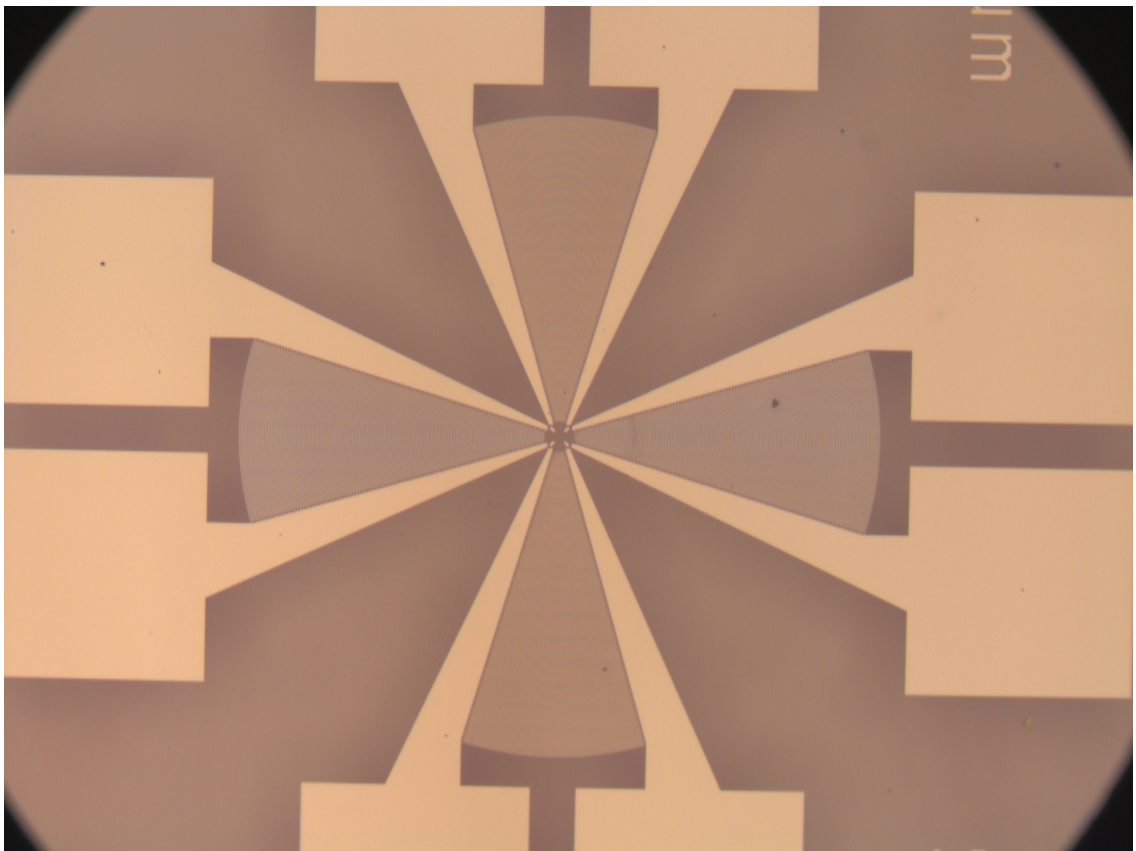


Figure 4.16: Close-up view of the 4 μm IDT cross.

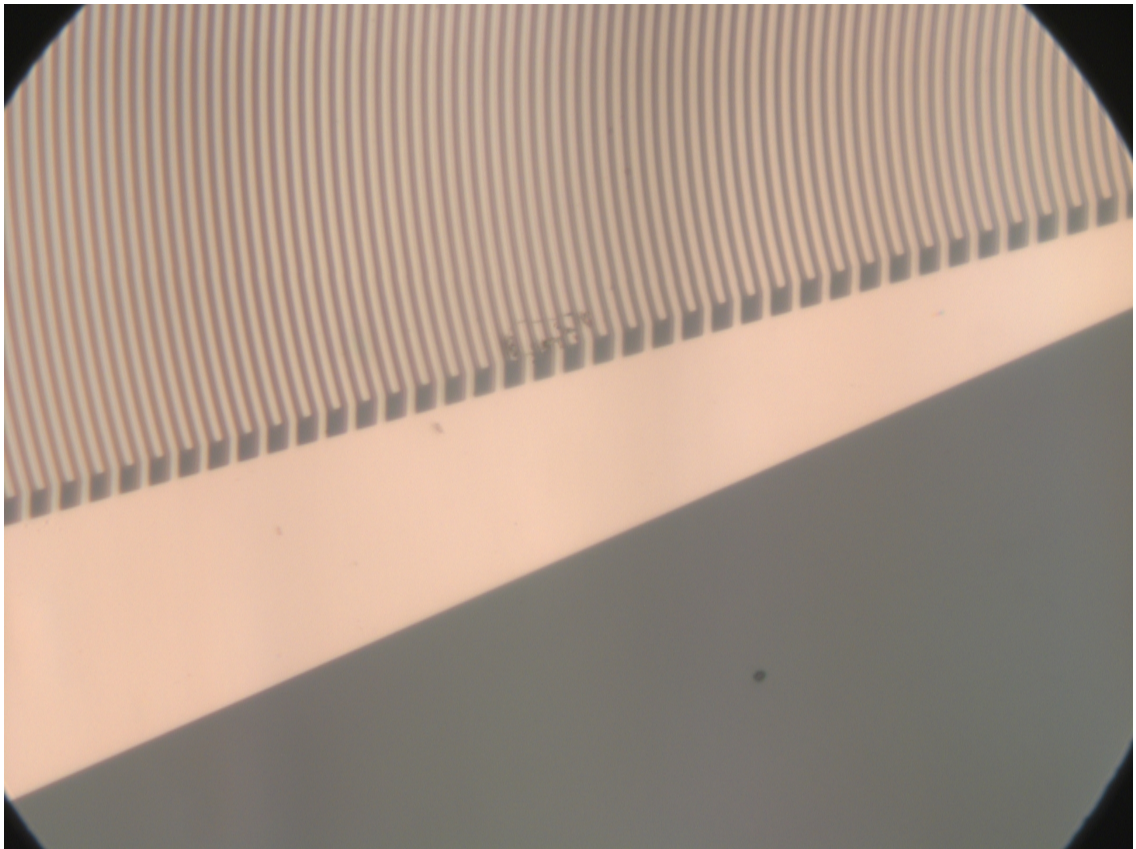


Figure 4.17: Close-up view of the 4 μm IDT fingers.

4.4 IDT Characterization

4.4.1 Vector Network Analyser and S-Parameters

Before presenting the frequency response measurements, it is useful to introduce the measurement tool and the measured quantity. The measurements were conducted using a vector network analyser, usually abbreviated as VNA. A VNA is used to measure the scattering parameters of RF and microwave networks. It measures the magnitude and phase of waves which are transmitted through and reflected from the device under test. Therefore, this tool is suitable for our measurements, since the IDTs are high-frequency structures and their response depends strongly on the applied frequency [64, p. 188].

The measured quantities are called scattering parameters, or S-parameters. In microwave network analysis, direct voltage and current measurements become difficult at high frequencies. Therefore, it is more useful to describe the network in terms of incident, reflected, and transmitted waves. Pozar explains that the scattering matrix relates the voltage waves incident on the ports to the voltage waves reflected from the ports [64, Sec. 4.3]. For a two-port network, the most relevant parameters are S_{11} , S_{21} , S_{12} , and S_{22} .

Here, S_{11} describes the reflection coefficient seen at port 1, while S_{21} describes the transmission coefficient from port 1 to port 2. In our measurements, S_{11} is mainly used to observe how much of the signal is reflected from one IDT, and therefore how well the IDT is matched at a certain frequency. On the other hand, S_{21} is used to observe how much signal is transmitted from one IDT to the other. Since these parameters are measured as a function of frequency, they allow us to determine the frequency response of the fabricated IDT structures [64, Sec. 4.3].

4.4.2 Frequency Response Measurements

For the frequency response measurements, we first estimated the expected resonance frequencies of the IDTs from the relation

$$f = \frac{v}{\lambda}. \quad (4.1)$$

A surface acoustic wave velocity of $v = 4888$ m/s was used, based on previous measurements conducted on ZnO/SiC samples [37]. Therefore, the expected frequencies for the three IDT wavelengths are

$$\begin{aligned}\lambda = 4 \mu\text{m} &\Rightarrow f = \frac{4888}{4 \times 10^{-6}} = 1.222 \times 10^9 \text{ Hz} = 1222 \text{ MHz}, \\ \lambda = 6 \mu\text{m} &\Rightarrow f = \frac{4888}{6 \times 10^{-6}} = 8.147 \times 10^8 \text{ Hz} = 814.7 \text{ MHz}, \\ \lambda = 8 \mu\text{m} &\Rightarrow f = \frac{4888}{8 \times 10^{-6}} = 6.11 \times 10^8 \text{ Hz} = 611 \text{ MHz}.\end{aligned}$$

We took these frequencies as the basis to set up our measurement devices. The frequency response measurements were conducted in the range of 400 MHz to 2200 MHz with 4000 points. The observed frequencies of Rayleigh and Sezawa modes were close to the expected frequencies. In the graphs, the measured signals are shown through the S-parameters S_{11} , S_{21} , S_{12} , and S_{22} . As explained before, the reflection parameters S_{11} and S_{22} show the reflected signal at port 1 and port 2, while the transmission parameters S_{21} and S_{12} show the signal transmission between the two ports.

We conducted measurements for each pair of counter-facing IDTs. For each delay line, one IDT was connected to port 1 and the other to port 2, giving a total of 12 measured pairs. The IDTs with the same wavelength showed similar and functional behaviour. Therefore, only three representative measurements are shown below.

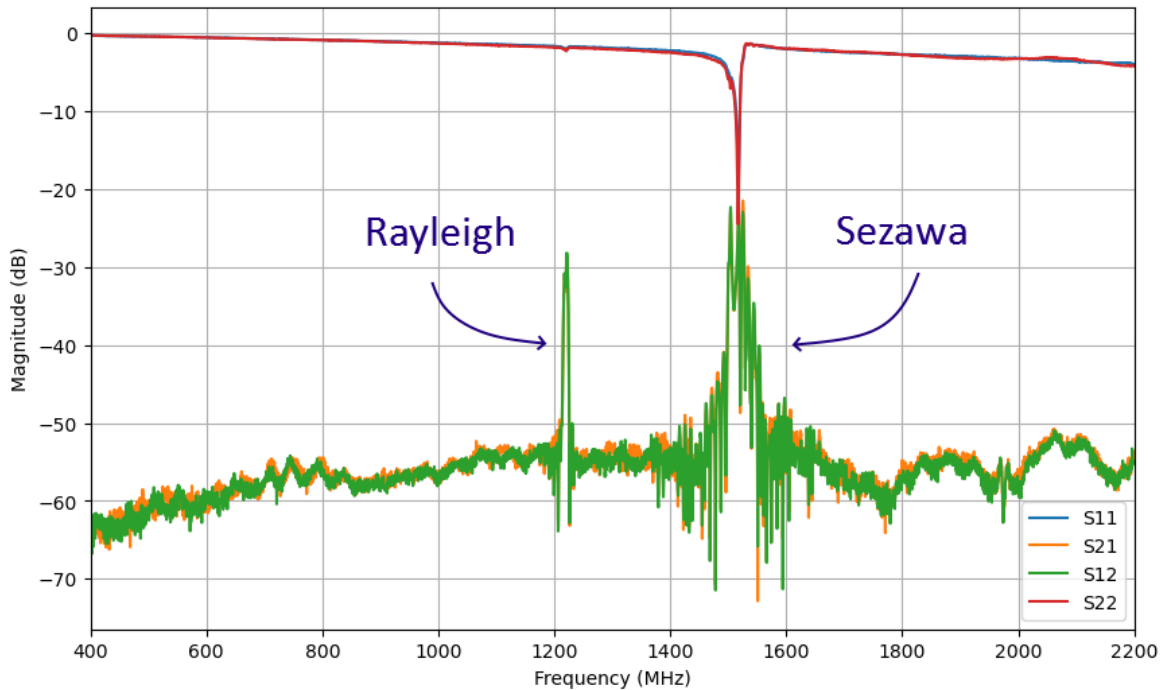


Figure 4.18: Frequency response of the 4 μm IDT, showing the Rayleigh and Sezawa modes.

In conclusion, the laser lithography and metal evaporation processes were successfully

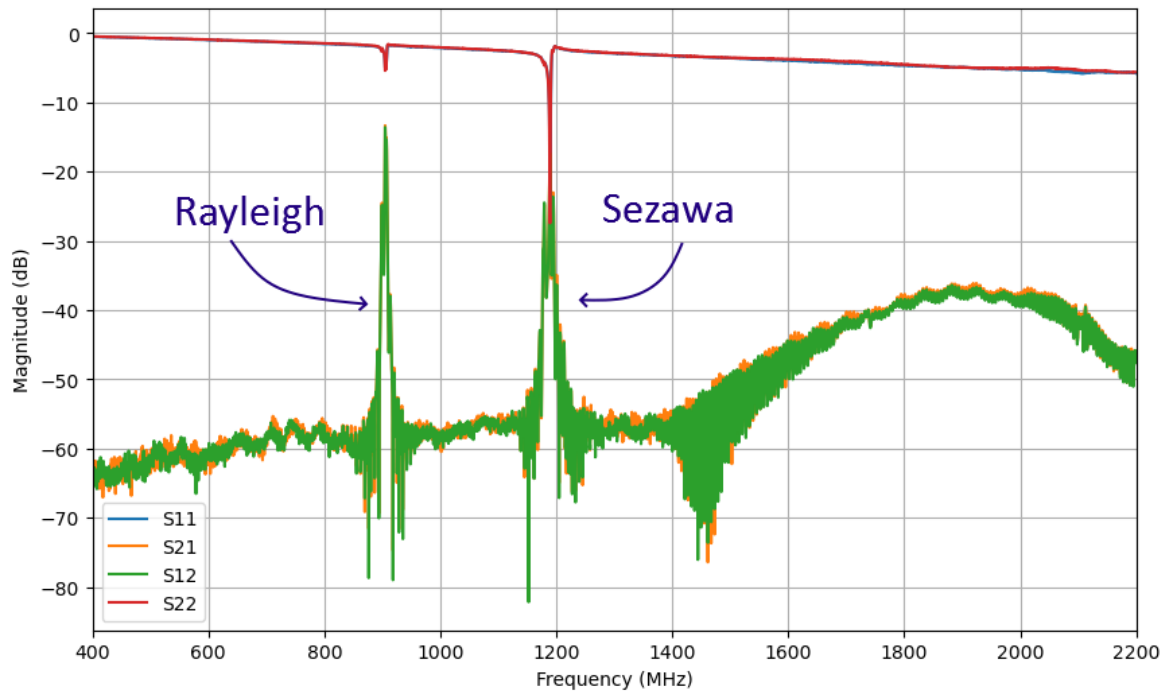


Figure 4.19: Frequency response of the 6 μm IDT, showing the Rayleigh and Sezawa modes.

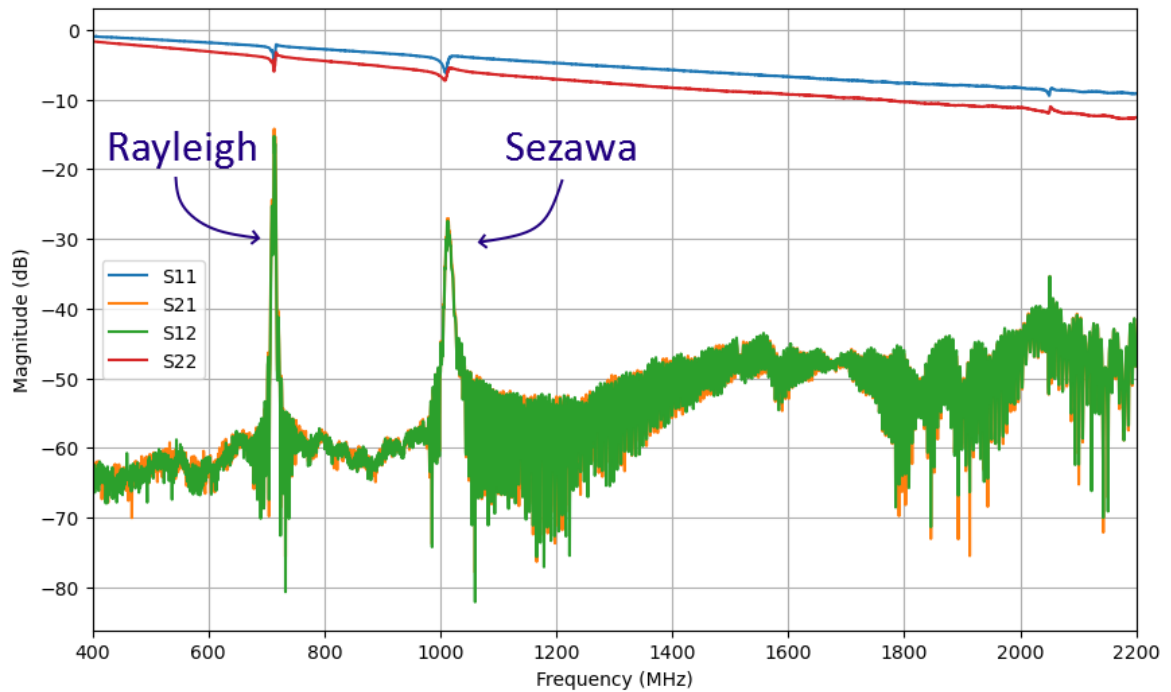


Figure 4.20: Frequency response of the 8 μm IDT, showing the Rayleigh and Sezawa modes.

implemented to fabricate functional IDTs on the substrate. This is supported by the frequency response measurements, where clear resonances were observed close to the expected frequencies for all tested wavelengths. The similar behaviour across multiple IDT pairs further supports the reliability of the fabrication process.

Although minor surface defects and contamination were observed during the optical inspection, these did not significantly affect the performance of the devices. This indicates that the overall process is robust. Nevertheless, further optimisation of the cleaning and fabrication steps could improve the device quality.

As the next step, we will carry out interferometry measurements to map the surface acoustic wave propagation and further analyse the IDT setup. Afterwards, we will conduct ODMR measurements coupled with PL measurements in order to detect the response of individual colour centres, which was theoretically discussed in the previous chapters.

4.4.3 Wiring of the Sample onto the Connector

On our connector chip, we have four ports for RF connection and four ports for DC connection. The long connection strips on the connector chip are used for RF, while the shorter, square-like pads are used for DC. The latter is not necessary for our purpose.

Due to the limited number of RF ports, we wired the counter-facing IDTs from one axis of the 6 μm and 8 μm quadra-pairs. For the initial tests, single wire bonding was used for all RF ports. After the tests, we observed that single wire bonding was not sufficient and did not provide a reliable connection. Many of the ports showed no resonant RF signal. One of the pairs worked well at the beginning. However, after some time, we observed burn marks on the metal pads of the 8 μm IDT, most likely caused by the high RF power. Therefore, we re-bonded the burned IDT pads and added extra wires, resulting in a double wire bond for each metal pad and improving the connectivity.

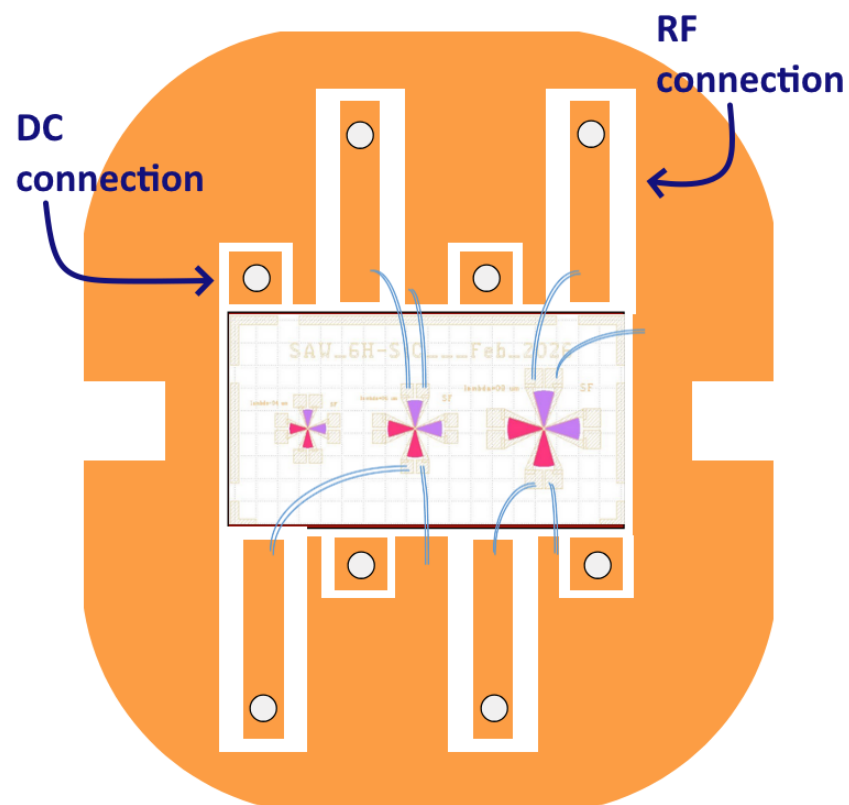


Figure 4.21: Wiring diagram of the chip.

4.5 Interferometry Mapping

4.5.1 Michelson Interferometry

We use a Michelson interferometry setup to characterise the SAW generation and propagation, as shown in Figure 4.22.

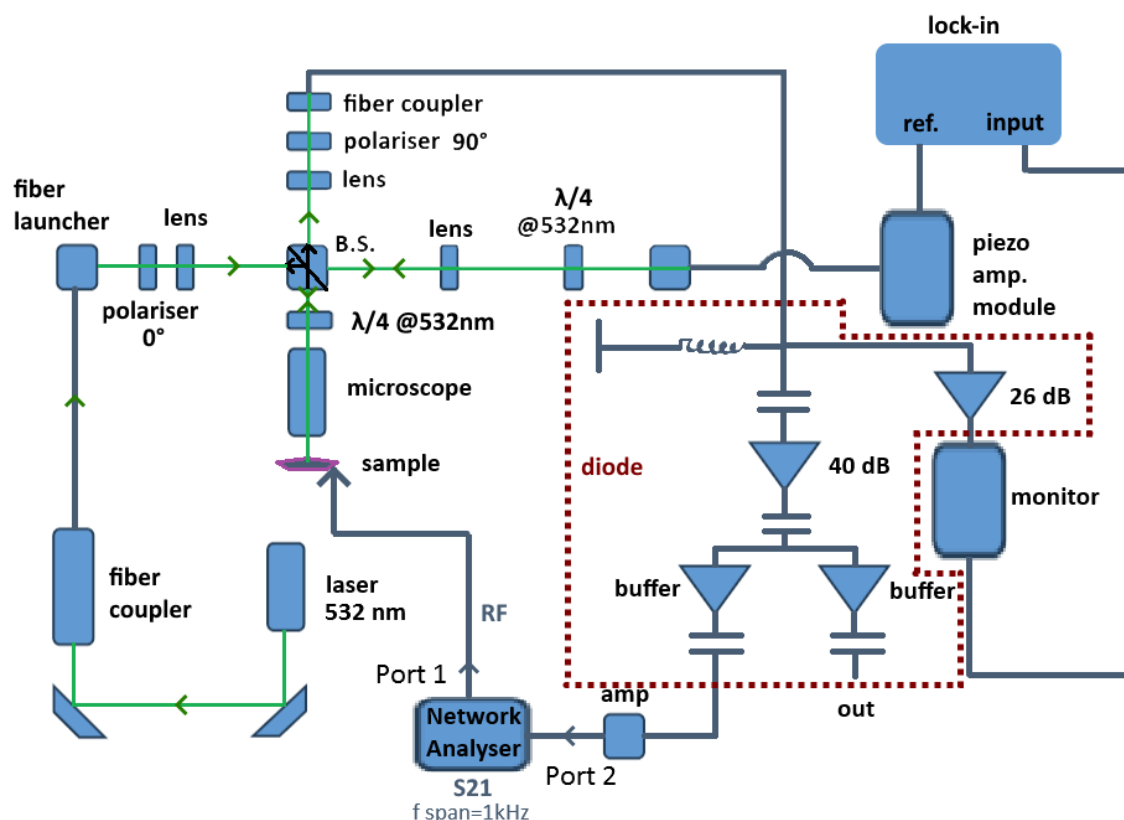


Figure 4.22: Michelson interferometer setup for surface vibration measurements on the sample, including reference arm phase modulation and amplified signal detection.

A Michelson interferometer-based optical measurement system is used to probe surface vibrations at the sample. In this method, small changes in the vertical surface displacement lead to changes in the optical path length. These changes are converted into phase changes of the reflected light and then into measurable intensity variations by interference [65]. A continuous-wave laser source is directed through a series of lenses for beam shaping and alignment, and through a polariser set to 0° to define a clean linear input polarisation. The beam then reaches a beam splitter (B.S.), which acts as the central optical element of the setup. From the B.S., the light is directed towards the sample path and the reference path with the piezo-controlled mirror. The returning light from these paths is recombined at the

B.S. and then directed towards the detection path.

In the sample path, the beam is directed from the B.S. towards the sample. It passes through a quarter-wave plate ($\lambda/4$ at 532 nm), then through the microscope objective, and is focused onto the sample surface. In this way, the setup can probe the surface displacement caused by the SAWs. The reflected light from the sample passes through the same quarter-wave plate again and returns to the B.S. Due to this double pass through the quarter-wave plate, the polarisation of the returning light is rotated by 90° with respect to the incoming light. The reflected light carries phase information related to the surface displacement.

In the reference path, the beam travels from the B.S. towards the piezo-controlled mirror. It passes through another quarter-wave plate ($\lambda/4$ at 532 nm) and is reflected back from the mirror mounted on the piezoelectric actuator. The piezo amplifier module drives this mirror and therefore modulates the optical path length of the reference path. This modulation is used as the reference signal for the lock-in amplifier, enabling phase-sensitive detection.

The light returning from the sample path and the reference path recombines at the B.S. The recombined light is then selected by the polariser set to 90° and sent through the fibre coupler into the detection path. In the detection path, the interference signal reaches the photodiode. This signal is then amplified using the 40 dB and 26 dB gain stages before being sent to the monitor and the lock-in amplifier. In this way, small phase variations caused by the SAW-induced surface motion can be measured.

At the same time, one of the interdigital transducers (IDTs) is connected via an RF line to the network analyser. The applied RF signal excites SAWs on the sample through piezoelectric coupling, where the periodic electric field generated by the IDT produces acoustic waves on the substrate surface [45]. By combining the RF excitation with the interferometric detection, we can gain information on the phase and amplitude of the surface motion relative to the resonance frequency of the IDT. In other words, the RF is sent to the IDT via port 1 and the output of the photodiode goes to port 2. By measuring S_{21} as a function of position, one gets the spatial map. The resulting surface, phase, and amplitude maps are discussed in Section 4.5.2.

4.5.2 2D Surface Scans

In Figure 4.23, we see the centre surface of the $8\ \mu\text{m}$ IDT. It was scanned using the interferometry setup in order to map the surface. This measurement took 27 h in total, with 121 cycles on the x axis and 141 points on the y axis.

We can clearly observe some distortions in the SAW map in Figure 4.24. Since the same periodic distortions are also visible in Figure 4.23, we can conclude that this is not due to the wave nature itself, but rather due to an experimental artefact. We suspect that it may be

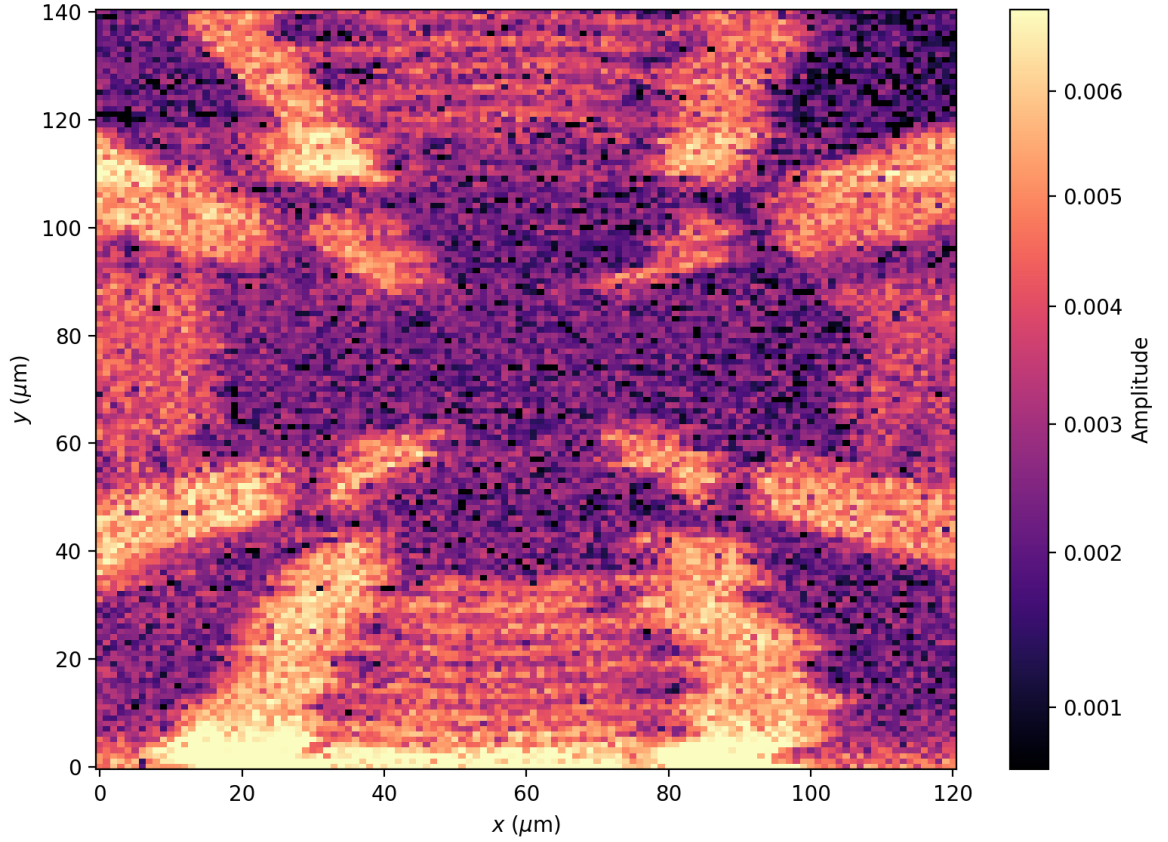


Figure 4.23: Surface scan of the 8 μm IDT centre.

caused by the imperfect reliability of the motors during extended run times.

Nevertheless, one can still observe some important features. We see that the excited IDT on left side generates SAWs, and these SAWs propagate along the x direction towards the counter-facing IDT, as shown in Figure 4.25. We can also observe that the waves successfully focus at the centre.

4.5.3 Frequency Response Measurements

In Figures 4.26 and 4.27, we measured the resonance frequencies of the IDTs by applying an excitation frequency of 713 MHz. Due to mass-loading effects caused by the metal IDTs, these resonance frequencies may be slightly shifted with respect to the regions without metal layer on top of the ZnO/SiC [66]. Therefore, we conducted multiple measurements in order to investigate this behaviour.

We compare the S_{21} resonances of the IDTs with Michelson interferometer measurements which were measured from the centre of the delay lines. From our measurements we know

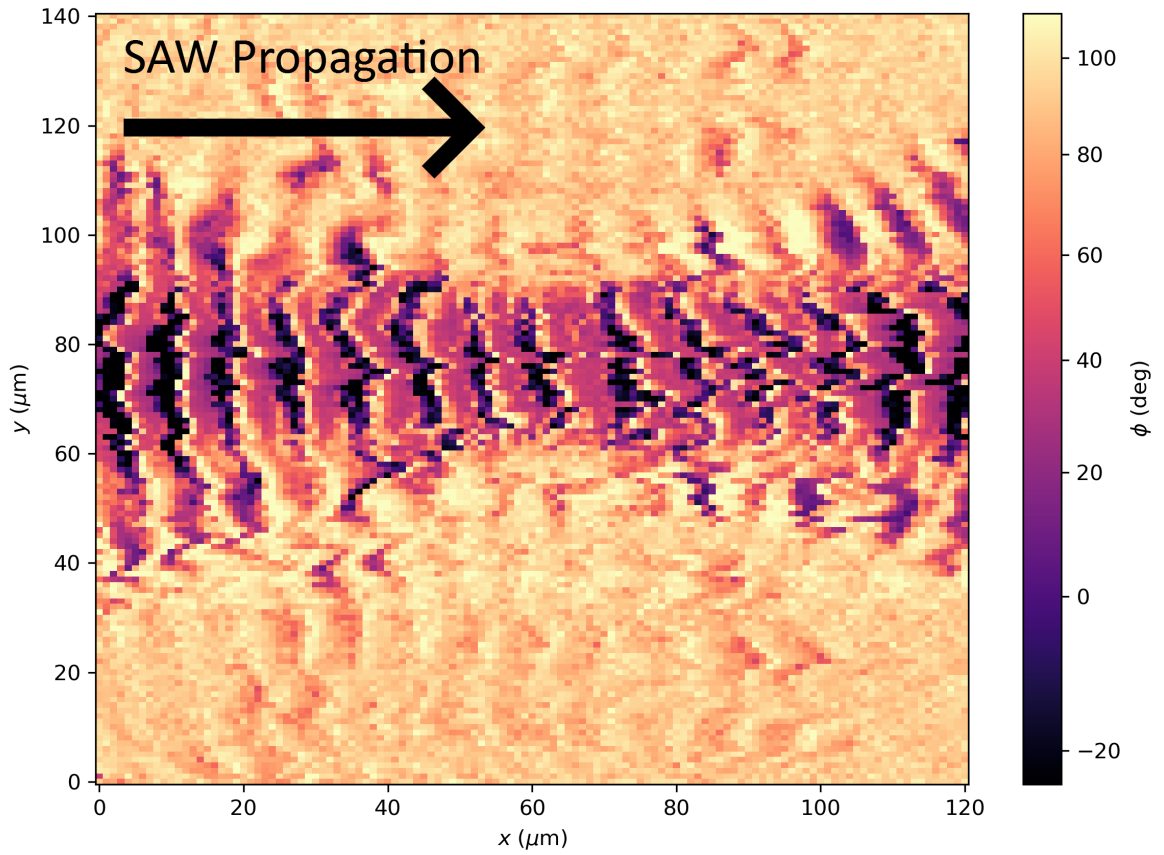


Figure 4.24: Phase map of the surface scan at the 8 μm IDT centre.

that the dominant resonance frequencies of the 4 μm IDT are 1221 MHz for the Rayleigh mode and 1525 MHz for the Sezawa mode, with the latter having the higher amplitude. For the 8 μm IDT, the dominant resonance frequencies are 714 MHz for the Rayleigh mode and 1013 MHz for the Sezawa mode, with the Rayleigh mode having the stronger amplitude. Rayleigh and Sezawa modes are both acoustic modes that can appear in layered structures, and they can show different coupling strengths and sensitivities depending on the substrate and the geometry [67].

In Figure 4.26, we can see that the Rayleigh resonance observed in S_{21} around 1221 MHz is also visible in the Michelson measurement, but it has a rather weak signal compared to the neighbouring periodic resonances. Around the expected Sezawa resonance region, the S_{21} signal shows a strong response, while the Michelson signal does not show a corresponding strong peak. Instead, the Michelson measurement is dominated by several periodic resonances.

In Figure 4.27, we can see that the Rayleigh resonance observed in S_{21} agrees well with

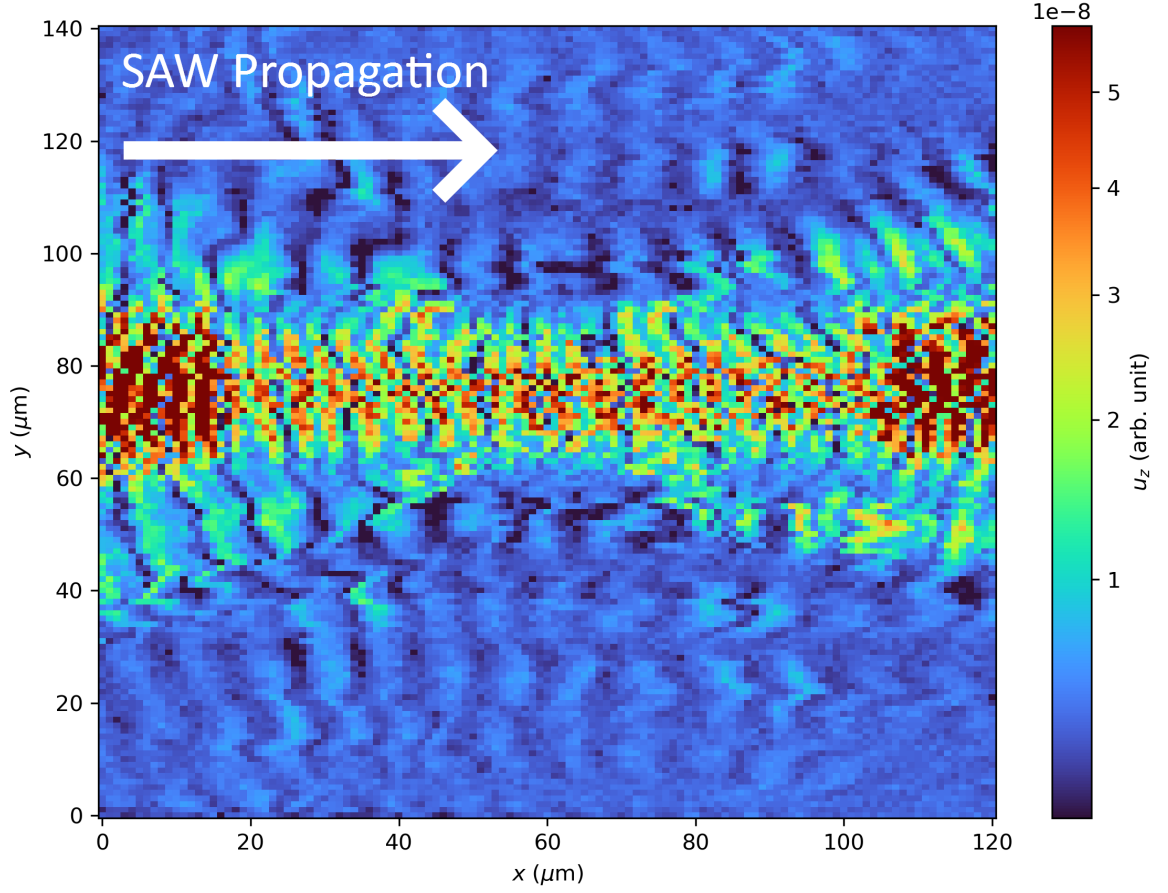


Figure 4.25: Amplitude map of the surface scan at the $8 \mu\text{m}$ IDT centre.

the Michelson measurement. More interestingly, close to this resonance, we observe a strong peak in the amplitude of the Michelson interferometer signal. Since similar periodic features are visible across the measured frequency range, and since their amplitudes depend on the measurement position, we attribute these resonances to bulk acoustic waves excited by the IDT out of their SAW resonance frequencies and reflected from the bottom of the substrate. Bulk acoustic waves can propagate through the substrate volume, in contrast to SAWs, which are mainly confined close to the surface [68]. Bulk acoustic wave radiation and related substrate effects can also appear in SAW resonator measurements [69].

As the angle of incidence of these reflected waves changes, their amplitude also changes. This explains why the amplitude of the periodic resonances is position-dependent, particularly along the x axis.

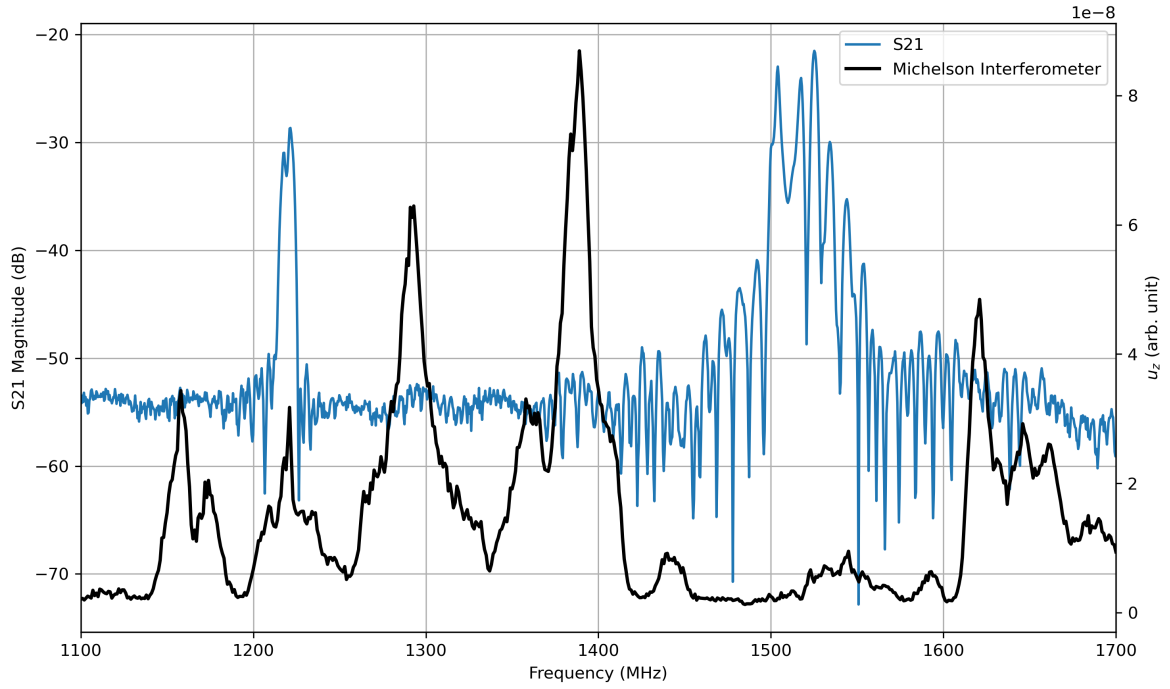


Figure 4.26: Frequency response of SAWs excited by the IDT with $\lambda = 4 \mu\text{m}$, measured in the range of 1100-1700 MHz with 101 points, averaging number of 4, and RF power of 0 dBm.

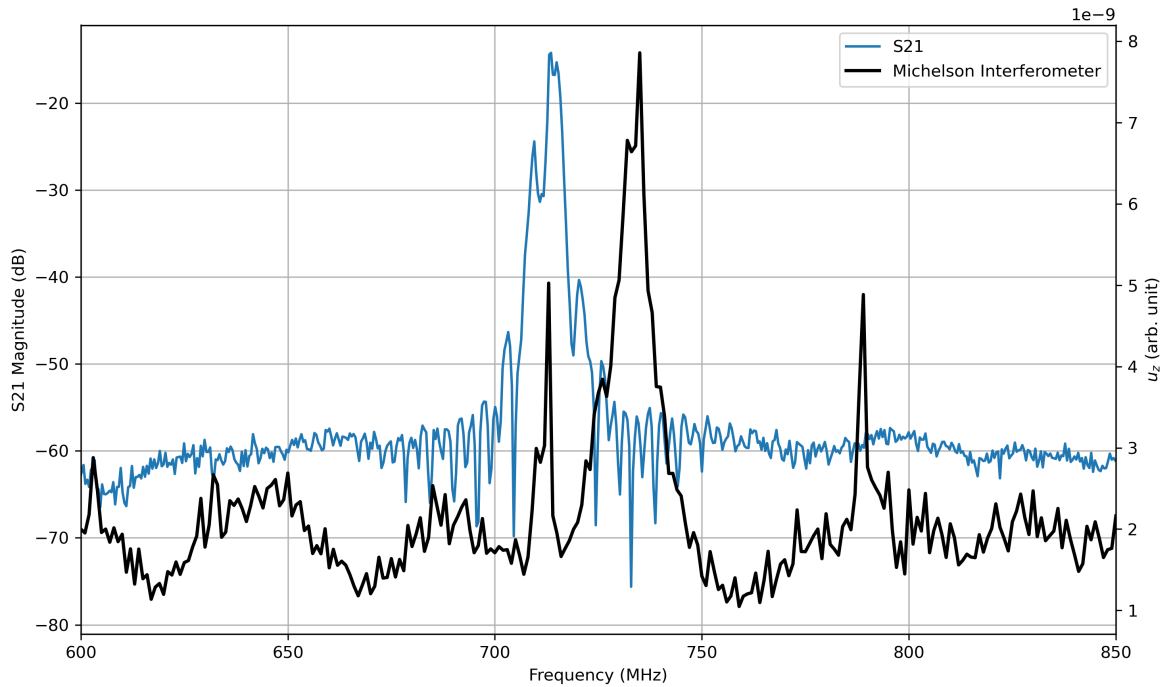


Figure 4.27: Frequency response of SAWs excited by the IDT with $\lambda = 8 \mu\text{m}$, measured in the range of 600-850 MHz with 101 points, averaging number of 4, and RF power of 0 dBm.

4.5.4 ODMR

In Figure 4.28, one can see the optically detected magnetic resonance (ODMR) setup. We use a 532 nm Verdi laser to pump the Ti:Sapphire oscillator, which then provides the 780 nm excitation beam. After this line, the beam passes through adjustable filters, filter I and filter II, where we can change the attenuation during the alignment and the measurement process.

A chopper modulates the optical excitation at a fixed reference frequency. The chopper reference is sent to lock-in B, allowing the lock-in amplifier to selectively detect the optical signal component at that frequency and suppress low-frequency drift, laser intensity fluctuations, and broadband noise. After the chopper, the light passes through an 800 nm short-pass filter. This filter transmits the excitation light while blocking longer-wavelength background emission and infrared stray light. The 805 nm dichroic mirror then separates the excitation and detection paths by reflecting the excitation light towards the sample, while transmitting the near-infrared PL emitted from the sample.

On the left-hand side of the setup, the excitation light is focused onto the sample through a 50× objective with a numerical aperture of $NA = 0.55$. It is a confocal setup which gives a good resolution in the z direction. The sample is placed between the poles of a magnet, which allows us to tune the spin transitions by changing the external magnetic field. A camera is also included in the optical path in order to monitor the sample position and assist the alignment.

The returning light carries the PL emitted from the sample. This PL passes through the 805 nm dichroic mirror and then through the 850 nm long-pass filter. The 850 nm filter spectrally isolates the desired near-infrared emission from the sample and suppresses residual laser light and out-of-band background. This improves the contrast and signal-to-noise ratio of the ODMR signal. The filtered PL is then focused onto the diode and sent to the lock-in amplifiers.

From our frequency response measurements, we determine the Rayleigh frequency of the sample and use this frequency to drive the RF generator. The RF signal is amplitude-modulated (AM) at 404 Hz. This modulation frequency is used as the reference input for lock-in A. The output of the RF generator is amplified and sent to the IDT on the sample. Thus, the IDTs are excited and SAWs are generated. A reference signal from the signal generator is also sent to lock-in A, so that the SAW-induced modulation of the PL can be detected phase-sensitively.

When we excite the IDT at the Rayleigh frequency, the generated SAWs create strain fields on the sample. These strain fields can couple to the spin states through spin-phonon interaction. By changing the magnetic field, we can select which spin transitions are expected to occur. During this process, the population of the spin states changes, which leads to a change in the emitted PL intensity. Therefore, the detected PL contains information about

the spin-state population.

Lock-in B records the optically modulated PL signal, while lock-in A measures the variation related to the RF-driven SAW excitation. Therefore, by comparing the signals from lock-in A and lock-in B, we can observe small variations in the spin-state population while reducing the effect of laser intensity fluctuations. By comparing the observed peaks with our theoretical calculations, we can identify which transitions occur at which magnetic fields in the spectrum and determine the individual colour centres.

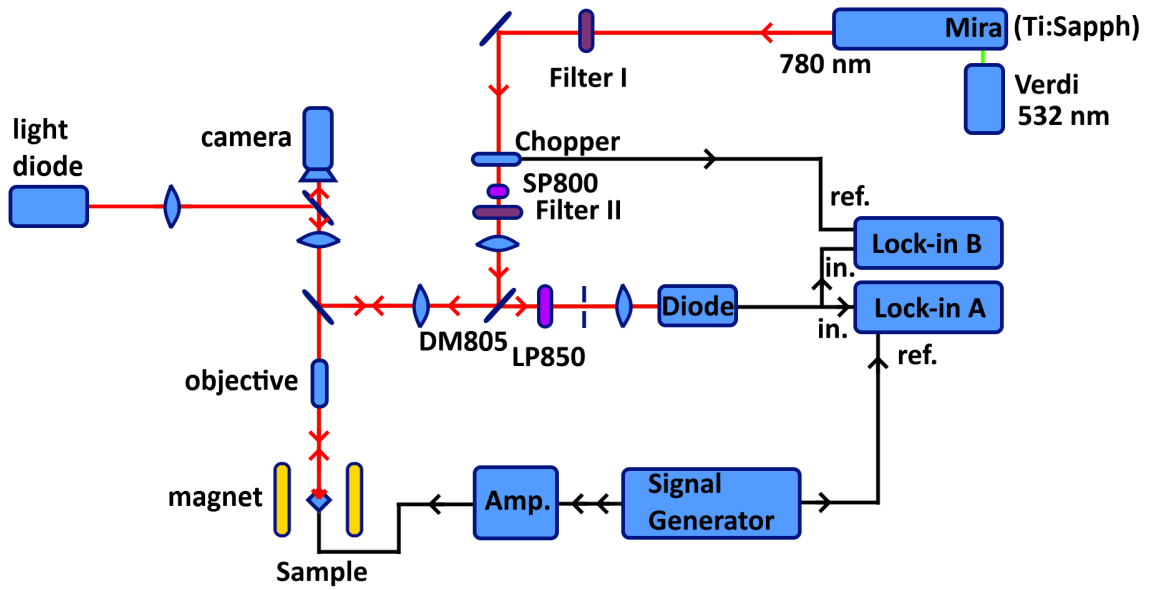


Figure 4.28: ODMR setup.

We conducted numerous ODMR measurements on the $6\ \mu\text{m}$ and $8\ \mu\text{m}$ IDTs. During these measurements, the IDTs were driven at their resonance frequencies using RF signals, and the emitted PL was recorded while changing the magnetic field. Throughout these measurements, we observed some notable signals which could be related to key colour centres. Nevertheless, due to the high noise level, we concluded that more extensive measurements are required.

The measurements were conducted at room temperature with an integration time, meaning the measurement time for each point, of up to 90 s. In our measurements, we mostly observe noise. In comparison to the measurements reported by Vasselon et al. [13], we were expecting a broad peak as shown in Figure 4.29. Although we can make a rough comparison, the measured signal is still dominated by noise. We suspect that this may be related to the short lifetime of the spin states of 6H-SiC at room temperature.

Although we conducted numerous measurements, here we show two of the most notable ones. The measurement in Figure 4.29 was conducted by exciting one of the $6\ \mu\text{m}$ IDTs in

the magnetic-field range of 0–60 mT. The measurement was conducted in two sessions, and in the graph these two data sets are shown together. Figure 4.30 shows the measurement conducted by exciting one of the 8 μm IDTs in the magnetic-field range of 15–40 mT.

During the measurements, we obtained the best results with longer measurement times. In this case, the integration time was 30 s per point. By comparing the peak positions in the graphs with the calculated transition strength as a function of magnetic field we see some rough signals which may be linked to these transitions.

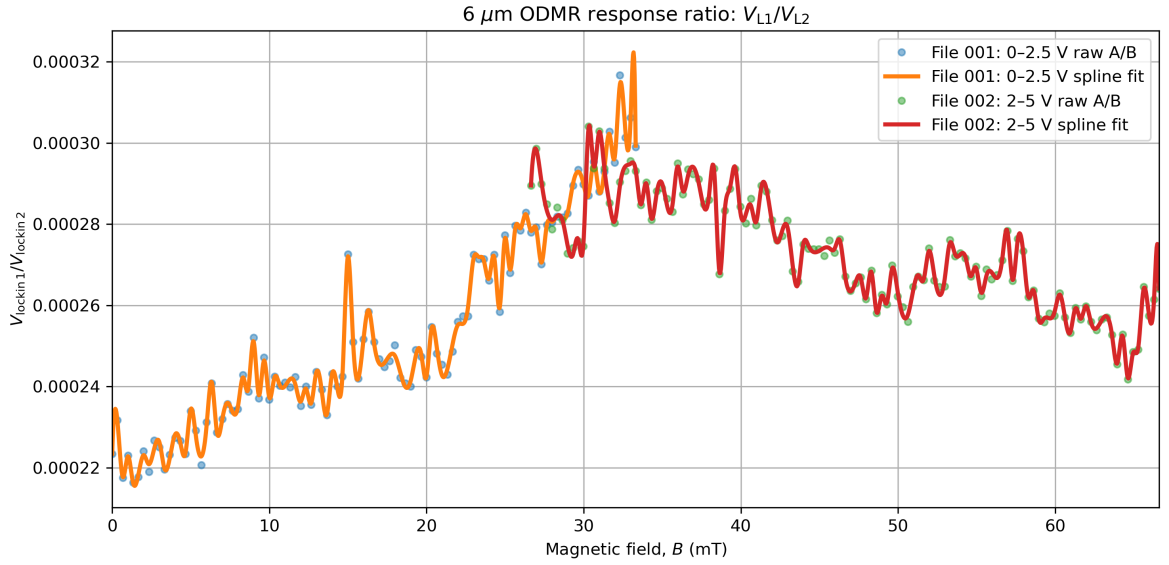


Figure 4.29: ODMR measurement on the 6 μm IDT, where the laser is pointed at the centre.

This observed behaviour is discussed in more depth in section 5.

Increasing the measurement time for individual points gave more distinguishable structures in the measured spectra. Therefore, longer scan times are expected to improve the quality of the ODMR measurements, especially when comparing different IDTs and different angles between the IDT axis and the magnetic field. One downside of this approach is that the measurements take several hours, which slows down the experimental process. For example, Ramsay et al. reported measurement times of up to two weeks for spin-relaxation measurements of qudit modes [70]. Unfortunately, our current laboratory setup does not allow such extended measurements.

Another option for improving the measurement quality is to cool down the substrate to cryogenic temperatures. At low T , phonon mediated spin relaxation processes can be reduced, which may result in a longer spin coherence time and improved visibility of spin related signals [71]. Separately, as discussed in Section 3.1, the zero-field splitting parameter can depend on temperature, which affects the spin-level structure of the colour centres.

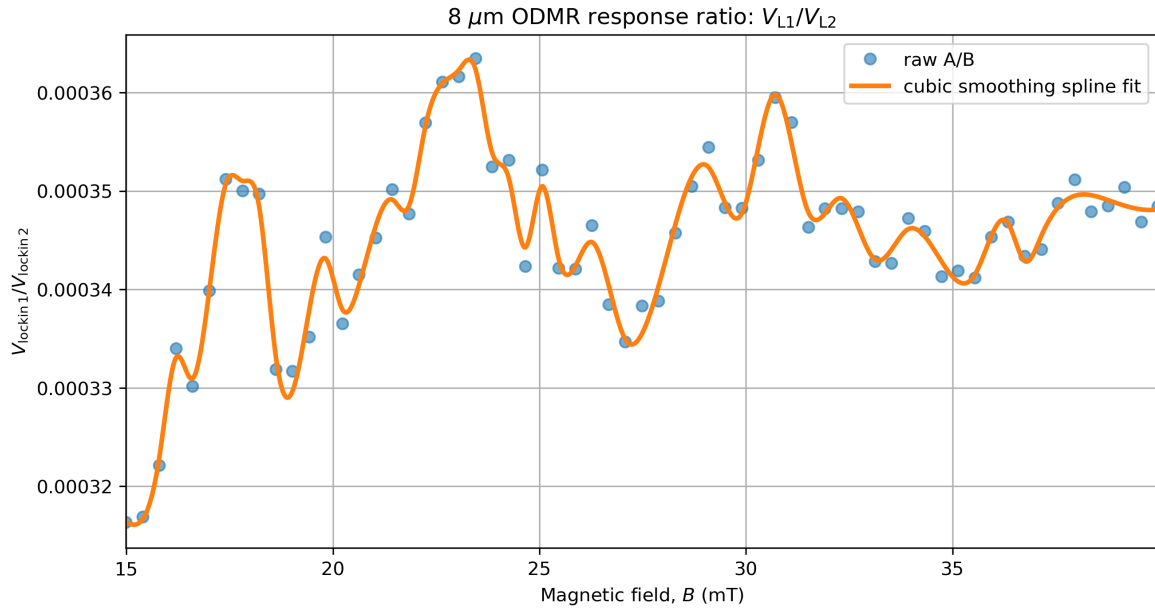


Figure 4.30: ODMR measurement on the 8 μm IDT, where the laser is pointed at the centre.

Therefore, this temperature dependence should also be considered when performing cryogenic measurements. However, with the current equipment, changing the angle of the magnetic field is not possible when the substrate is placed inside the cryogenic chamber. Therefore, despite these limitations, the main improvement route was to increase the scan time, while cryogenic measurements remain a possible next step.

Another issue was related to the long-term stability of the fabricated sample. Since the substrate with IDTs is very delicate, extended handling and exposure to unprotected air conditions over several months led to the accumulation of dust particles on the IDTs, affecting their performance. In the frequency response measurements, we observed that some of the IDTs lost their functionality, while the working ones showed poorer efficiency. This was also related to the wire bonding, as discussed previously. Since the bonding wires are very delicate, even small vibrations can cause them to break off.

Since we had a second substrate available, sample B, we applied the same nanofabrication techniques to obtain a fresh set of IDTs for further measurements. For this second substrate, the sputtering temperature was reduced to 200°C in order to improve the ZnO layer quality. As discussed in the sputtering section, the following X-ray scan and ω -scan measurements showed an improved crystalline quality of the ZnO layer, with the reduced rocking-curve width shown in Figure 4.9b. This result supports that lowering the sputtering temperature improved the layer quality and provided more efficient piezoelectric film for SAW excitation.

4.5.5 Cryogenic Cooling

In order to improve the ODMR measurements, one possible approach is to cool down the sample. As mentioned earlier, cooling down the sample can reduce phonon-mediated spin relaxation processes, which may improve the spin coherence time and the visibility of spin-related signals [71].

In our case, cryogenic cooling is achieved using a helium-based cooling system. Helium flows through a cooling tube inside the vacuum chamber, and the sample is cooled indirectly through thermal contact with this cold region. The investigated device is an IDT structure fabricated on a 6H-SiC substrate. The sample is positioned close to the centre of the chamber, while the helium tube passes nearby. Heat from the sample is conducted through the sample holder towards the helium tube, transferred through the tube wall, and carried away by the flowing helium. Thus, the cooling takes place via thermal conduction in a cold-finger cryostat.

Before helium cooling is started, the sample chamber is evacuated to a pressure of approximately 10^{-6} mbar. The vacuum reduces heat transfer from the outer walls of the cryostat towards the sample through the residual gas. Thermal radiation is minimized by the metal shield, while the remaining heat load mainly comes from solid conduction through the sample holder, electrical wires, and mechanical supports [72, 73]. Evacuating the chamber also prevents water vapour, nitrogen, and oxygen from condensing on the sample or the cooling tube.

After the required vacuum level is reached, the helium supply valve is opened gradually and the circulation pump is started. The helium remains separated from the vacuum space by the tube wall. As helium flows through the internal tube, heat is removed from the sample holder and the IDT sample. The cooldown rate depends on the thermal mass of the sample assembly, the thermal contact with the cooling tube, and the available helium cooling power.

The sample does not necessarily reach the same temperature as the helium because heat passes through the sample, the sample holder, their contact, and the tube wall. Therefore, the temperature sensor should be placed as close as possible to the IDT substrate. A sensor placed only on the helium inlet tube would mainly measure the helium temperature and may not represent the actual temperature of the device.

The cooling procedure starts by mounting the IDT sample in the centre of the vacuum chamber and checking the electrical connections, temperature sensors, helium cooling line, and sealing rings. The vacuum pump is then started while the helium valve remains closed. The chamber is evacuated until the pressure reaches approximately 10^{-6} mbar. If the required pressure cannot be reached, this may indicate a weak joint or a leak in the system. Once the vacuum becomes stable, the helium supply valve is opened gradually and the circulation pump is started.

Measurements should be started only after the sample temperature, vacuum pressure, helium pressure, and helium flow have stabilized. Our system is designed to operate around 20 K. For temperatures above 20 K, the integrated heater is activated to counterbalance the cooling effect. For temperatures below 20 K, the helium flow rate from the storage container is increased.

Weak thermal contact, heat conduction through the electrical wires, thermal radiation from the chamber walls, and insufficient helium flow can prevent the sample from reaching the expected temperature. Radiation shields and low-emissivity surfaces can be used to reduce the radiative heat load [72, 73].

The helium line contains refrigerated helium and pressurized flow components. Therefore, the cooling tube, valves, pump, and connections must be checked before operation. Helium should not be trapped between two closed valves because it expands strongly when it evaporates and warms up, which can cause a dangerous pressure increase. Since helium can also displace oxygen, the outlet and pressure-relief paths should be directed to a safe and ventilated location, and cryogenic personal protective equipment should be used [73].

The described cooling method is suitable for investigating the temperature dependence of the SAWs generated by the IDTs and the spin states in 6H-SiC. Since the sample is cooled indirectly, the sample temperature should be measured close to the device rather than inferred only from the nominal helium temperature.

4.6 Computation of the Data

With the conducted measurements we receive raw data. We need to process this data to transform it into information.

We recorded two lock-in signals, A_i and B_i , stored in the HDF5-formatted file. The ODMR response was normalised by dividing the fluctuating signal in channel A_i by the total photoluminescence intensity in channel B_i , thereby correcting for slow laser intensity fluctuations during the scan.

$$y_i = \frac{A_i}{B_i + \epsilon} \quad (4.2)$$

We set $\epsilon = 10^{-12}$. This is to prevent division by zero.

For the x-axis we have our range in magnetic field B (mT). But to reach here we first have to do a conversion. The change is being conducted on the voltage applied to the electro-magnet. For N data points, with $i = 1, \dots, N$, we have:

$$\Delta V = \frac{V_{max} - V_{min}}{N - 1} \quad (4.3)$$

$$V_i = V_{min} + (i - 1)\Delta V \quad (4.4)$$

Then we convert the voltage to the magnetic field with the following calibration factor:

$$x_i = B_i = V_i \cdot 13.3228 \frac{mT}{V} \quad (4.5)$$

Thus we have the data points:

$$(x_i, y_i) \quad (4.6)$$

This covers the plotting of the raw data. But now we also would like to fit this plot to have a smooth function, which is easier to read.

Smoothing splines were introduced in the classical work of Reinsch [74] as a method for constructing a smooth curve from noisy observations. In this thesis, the mathematical background is described using the penalised least-squares formulation given by Green and Silverman [75], while the numerical implementation is performed using SciPy's `UnivariateSpline` [76].

To obtain a smoother representation of the measured data, we apply a cubic smoothing spline. The idea can be explained step by step as follows. A spline is a piecewise polynomial function. In the cubic case, each piece is a polynomial of degree at most three. The spline finds a smooth function $f(x)$ where $f(x_i) \approx y_i$. Between two points t_j and t_{j+1} the function

is:

$$f_j(x) = a_j x^3 + b_j x^2 + c_j x + d_j \quad (4.7)$$

for

$$t_j \leq x \leq t_{j+1} \quad (4.8)$$

Here, the points t_j are called knots. A knot is a point where two neighbouring spline pieces meet. Thus the full fit can be written as:

$$f(x) = \begin{cases} f_1(x), & t_1 \leq x < t_2 \\ f_2(x), & t_2 \leq x < t_3 \\ \dots & \\ f_m(x), & t_m \leq x \leq t_{m+1} \end{cases} \quad (4.9)$$

At each internal knot, the pieces are connected smoothly. It means the function itself, its slope and its curvature are continuous.

$$f_j(t_{j+1}) = f_{j+1}(t_{j+1}) \quad (4.10)$$

$$f'_j(t_{j+1}) = f'_{j+1}(t_{j+1}) \quad (4.11)$$

$$f''_j(t_{j+1}) = f''_{j+1}(t_{j+1}) \quad (4.12)$$

The fitting procedure wants to stay close to the data by minimizing the residual contribution

$$\sum_{i=1}^N (y_i - f(x_i))^2 \quad (4.13)$$

whilst keeping the line smooth. This smoothness is achieved by controlling the curvature.

$$\int [f''(x)]^2 dx \quad (4.14)$$

Following the penalised least-squares formulation of smoothing splines, the overall process can be written as [75]:

$$S(f) = \sum_{i=1}^N (y_i - f(x_i))^2 + \lambda \int [f''(x)]^2 dx \quad (4.15)$$

The fitted function is the function which minimises $S(f)$. If we increase λ , the fit becomes smoother and ignores more noise. If λ is decreased, the fit follows the data more closely.

For our plotting, we used SciPy's `UnivariateSpline` [76]. In our case, we used the

default spline degree $k = 3$, corresponding to a cubic spline. In this implementation, the smoothing is not controlled directly by λ . Instead, SciPy uses a smoothing factor s . The input data are the points (x_i, y_i) , and the fitted spline is chosen so that the weighted residual sum of squares satisfies the smoothing condition given in the SciPy documentation [76]:

$$\sum_{i=1}^N [w_i (y_i - f(x_i))]^2 \leq s \quad (4.16)$$

Here, w_i are weights assigned to the data points. Since no weights were specified in our code, SciPy uses:

$$w_i = 1 \quad (4.17)$$

Therefore, the residual condition becomes:

$$\sum_{i=1}^N (y_i - f(x_i))^2 \leq s \quad (4.18)$$

In our case, we used:

$$s = 10^{-10} \quad (4.19)$$

and therefore:

$$\sum_{i=1}^N (y_i - f(x_i))^2 \leq 10^{-10} \quad (4.20)$$

In practice, this condition is satisfied numerically within the tolerance used by SciPy's fitting routine [76]. This differs from the classical penalised least-squares formulation written above. The theoretical expression uses λ to directly control the trade-off between closeness to the data and smoothness. SciPy's `UnivariateSpline` instead uses the allowed residual error s . Smaller s forces the spline to follow the data more closely, while larger s allows a smoother curve [76]. Since $s = 10^{-10}$ is very small, the fitted curve follows the measured data very closely. In addition, SciPy internally determines how many knots are needed and where they should be placed. Therefore, the data points (x_i, y_i) and the smoothing factor s are provided as inputs, while the spline knots and the cubic pieces are determined by SciPy's internal fitting algorithm.

Chapter 5

Experimental Results and Discussion

On Figure 5.1 we observe the Zero-field splitting of the colour centres at room temperature. The sample is excited with microwaves (MW) by placing the sample onto a waveguide and the measurement was conducted from the surface of the 6H-SiC. By running a frequency scan between 0 - 200 MHz with $B = 0$ mT we are able to recreate the measurement of Breev et al.[1]. The measurement shows the inverted sign of 2D of V3 with respect to V2 in the ground state. as well as giving information on the ground-state zero-field splitting values which are -28 MHz for V3(V1) and 128 for V2 centres.

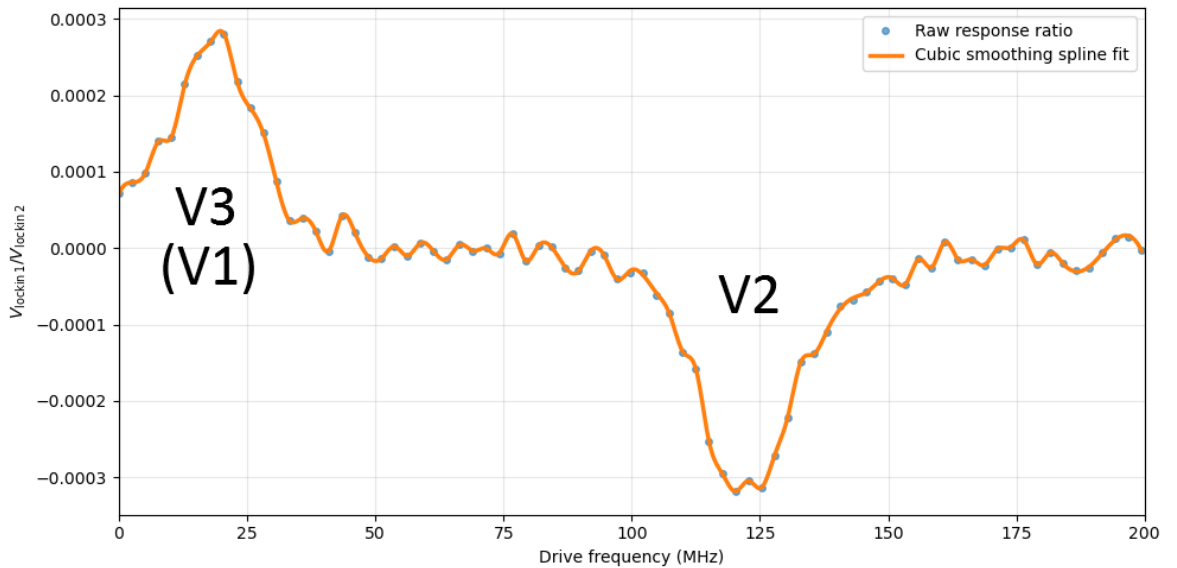


Figure 5.1: Frequency response measurement of the colour centres in 6H-SiC at room temperature and $B = 0$ mT over a frequency scan range of 0–200 MHz. The raw lock-in response ratio and the cubic smoothing spline fit are shown.

When we cool down the sample, the frequency response of the IDTs is not exactly the

same as at room temperature, and their acoustic resonances shift slightly towards a higher frequency. Therefore, before performing the ODMR measurements with SAWs, we run a frequency scan around the expected resonances studied in Section 4.4.2. This scan is performed in the ODMR setup at $B = 0$ mT, and the response is measured through changes in the photoluminescence. When the magnetic field and the SAW frequency are away from the acoustic-spin resonance, the generated SAWs can still modulate the detected PL through local vibrations at the sample surface. We use this effect to select the drive frequency that generates the strongest SAW response under the measurement conditions.

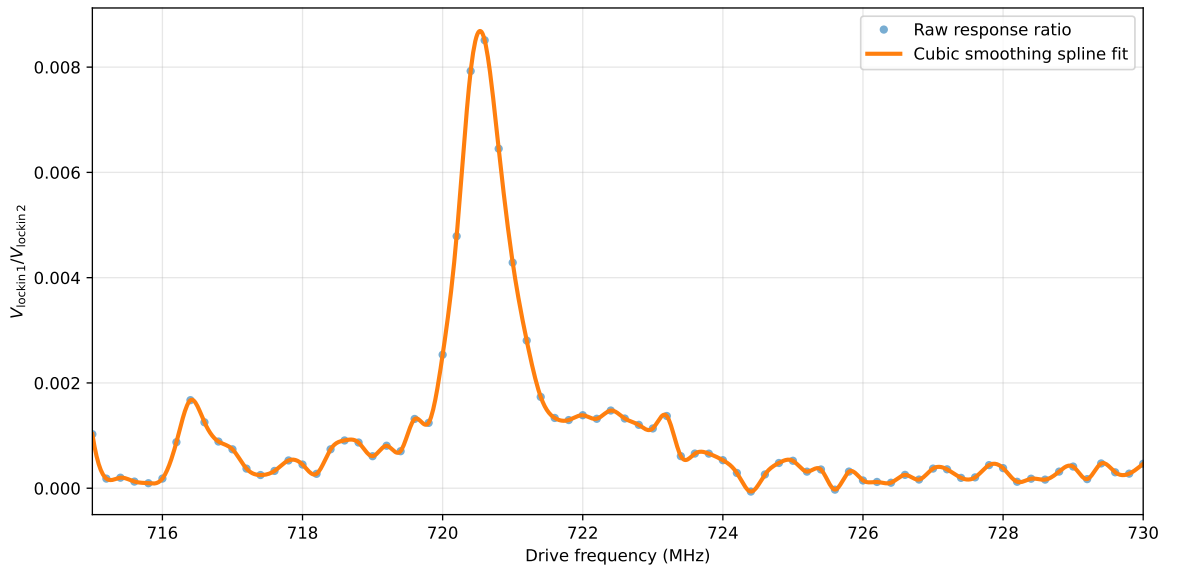


Figure 5.2: Frequency response measured through the PL in the ODMR setup at $T = 20$ K and $B = 0$ mT. The strongest response is observed around 720.3 MHz.

From the frequency response obtained in Figure 5.2, we set the drive frequency to 720.3 MHz and conducted ODMR measurements from 0 to 60 mT. We expected to see some apparent peaks which would have corresponded to different transitions in V2 and V3; GS and ES. On the Figure 5.3 and 5.4 we see two measurements conducted where the laser was pointed to the centre of the delay line with $8\mu\text{m}$ IDTs. Although, all the parameters of these consequent measurements were identical, the results are completely different. By repeating the ODMR measurements with different time scales, different ϕ_B , different temperatures with the $6\mu\text{m}$ and $8\mu\text{m}$ IDTs we couldn't get any repeatable measurements. We also tried, instead of measuring over and full range, 0-60 mT, we divided the measurement into smaller ranges of a span of 5 mT and ensured the stability of the system parameters such as the laser alignment and focus which affected the power of the measured PL. We also chose some ranges of regions which we thought that there may exist some transitions. But even after the

repetition of the measurement in a small region with same parameters no measurement were repeatable.

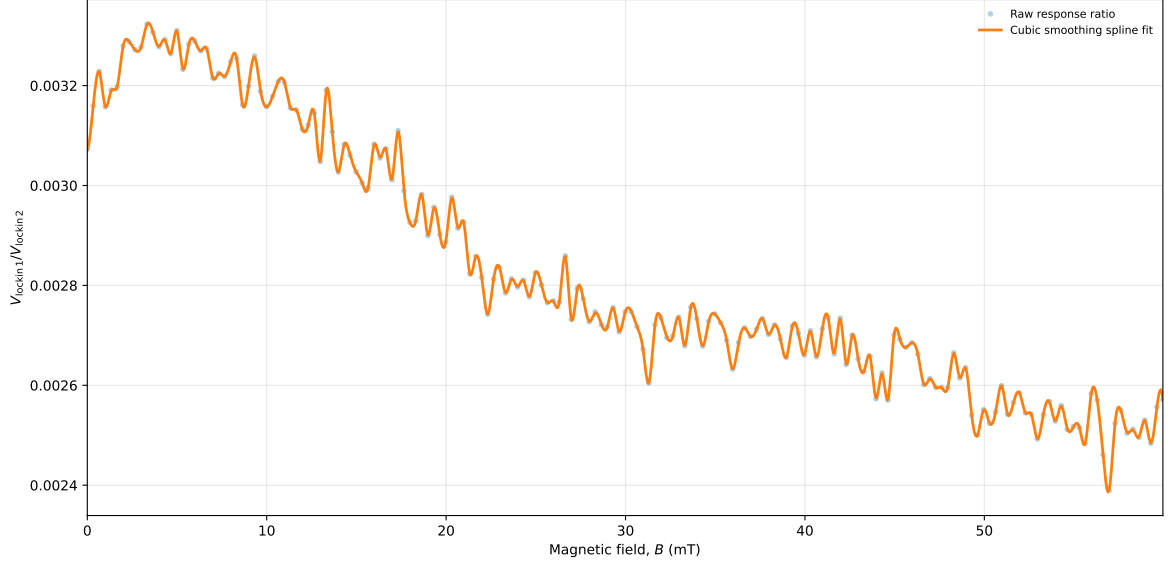


Figure 5.3: ODMR scan measured at $T = 20$ K using a drive frequency of 720.3 MHz over a magnetic field range of 0–60 mT, where the angle of the magnetic field with the SAW direction is $\phi_B \approx 45^\circ$. The field was scanned in steps of approximately 0.33 mT with an integration time of 10 s. The dots and the solid line represent the raw data and the cubic smoothing spline, respectively.

On Figure 5.5 we see an MW excited measurement where this time the magnetic field is kept constant at a comparatively lower magnitude of $B = 5.0243$ mT, while a frequency scan is conducted with the magnetic field oriented at $\phi_B = 0^\circ$ relative to the SAW direction. Although the measurement conducted with these conditions are also prone to the aforementioned problems, we can see some distinct peaks which overlaps with some of the expected transitions. For example, $V2\ 2 \rightarrow 3$, $\Delta m = \pm 1$ transition.

Overall, the ODMR measurements were not repeatable under our experimental conditions. Although some features overlapped with the calculated transition positions, these features could not be reproduced and therefore cannot be reliably assigned to specific spin transitions. Consequently, SAW-driven spin manipulation in 6H-SiC could not be experimentally confirmed in this work. We conclude that, on 6H-SiC, in our experimental configuration, the applied magnetic field reduces either the spin coherence time or the luminescence contrast between spin states to the extent that reliable ODMR measurements cannot be obtained.

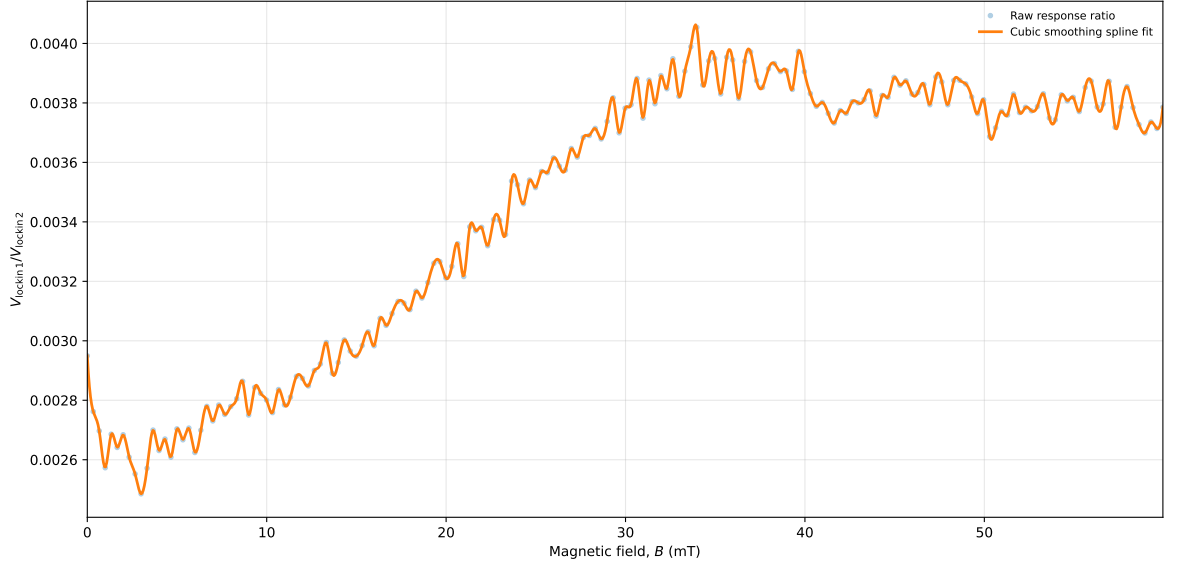


Figure 5.4: ODMR scan measured at $T = 20$ K using a drive frequency of 720.3 MHz over a magnetic field range of 0–60 mT, where the angle of the magnetic field with the SAW direction is $\phi_B \approx 45^\circ$. The field was scanned in steps of approximately 0.33 mT with an integration time of 10 s. The dots and the solid line represent the raw data and the cubic smoothing spline, respectively.

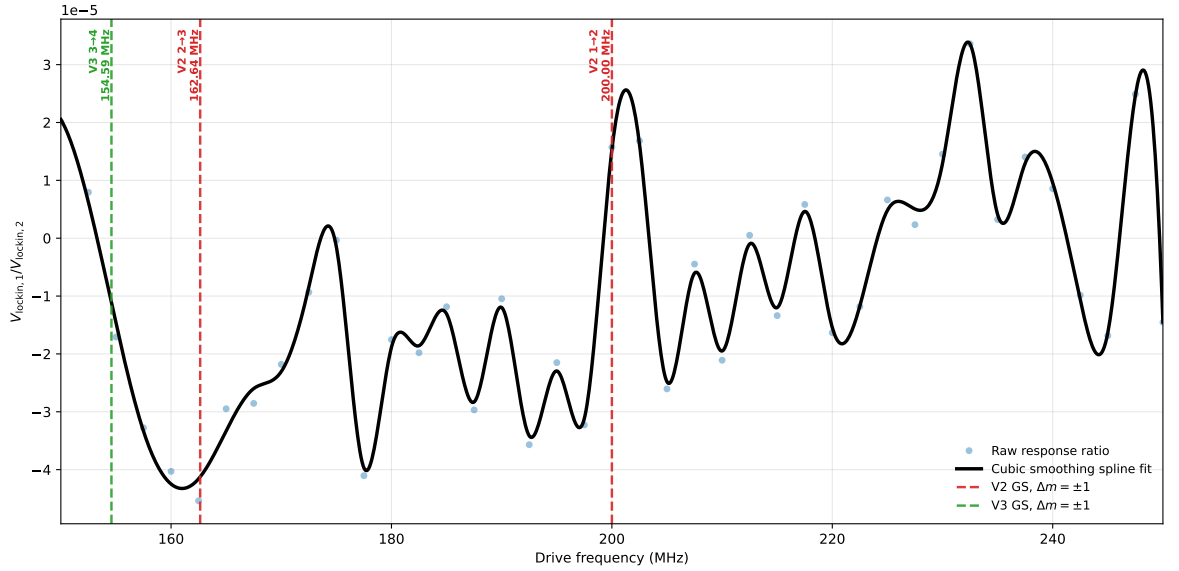


Figure 5.5: MW-excited frequency scan over the frequency range of 150–250 MHz at a fixed magnetic field of $B = 5.0243$ mT. The calculated transition lines correspond to ground-state (GS) transitions. A distinct peak overlaps with the expected V2 GS ($2 \rightarrow 3$) transition with $\Delta m = \pm 1$.

Chapter 6

Summary and Conclusions

The aim of this thesis was to investigate the acoustic manipulation of spin qubits in 6H-SiC by generating surface acoustic waves (SAWs) with interdigital transducers (IDTs). For this purpose, we studied the spin structure of the $V2$ and $V3$ silicon-vacancy centres, calculated the expected SAW-driven transition strengths, fabricated IDTs on ZnO/6H-SiC, characterised their acoustic response, and performed ODMR measurements under different experimental conditions.

From our theoretical calculations, we showed that the strengths of the SAW-driven spin transitions depend on the magnetic-field magnitude and its angle with the SAW propagation direction. We also compared the $\Delta m = \pm 1$ and $\Delta m = \pm 2$ transitions for the ground and excited states of the $V2$ and $V3$ centres. The calculations showed that the magnetic-field angle can be used to distinguish between different transitions. Based on the strain depth profile, we selected a depth of approximately 1000 nm for the creation of the silicon-vacancy centres.

The investigated substrate was confirmed as 6H-SiC by comparing its Raman spectrum with the reference spectrum. The silicon-vacancy centres were created by proton irradiation with an implantation energy of 175 keV. The measured photoluminescence spectra were compared with the expected emission from the colour centres. At cryogenic temperatures, the spectral features become more distinguishable, allowing the contributions from the different silicon-vacancy centres to be studied more clearly.

For the generation of SAWs, a ZnO layer was deposited on the 6H-SiC substrate, followed by the fabrication of IDTs with different wavelengths. The first ZnO layer had an ω -scan rocking-curve FWHM of 5.02° . For the second substrate, the sputtering temperature was reduced to 200°C , resulting in a reduced FWHM of approximately 2.7° . This indicates an improvement in the crystalline alignment of the ZnO layer.

The fabricated IDTs were characterised using frequency-response measurements. For the

4 μm IDT, the dominant Rayleigh and Sezawa resonances were observed around 1221 MHz and 1525 MHz, respectively. For the 8 μm IDT, these resonances were observed around 714 MHz and 1013 MHz. The Michelson interferometry measurements confirmed the generation and propagation of SAWs in the delay lines. From the spatial maps, we could also observe that the waves focus near the centre of the delay line. In addition to the SAW resonances, several periodic features were observed and attributed to bulk acoustic waves reflected from the bottom of the substrate.

At room temperature and $B = 0$ mT, we measured the zero-field response of the silicon-vacancy centres over the frequency range of 0–200 MHz. The observed responses were compared with the expected zero-field splitting values of the $V3$ ($V1$) and $V2$ centres.

We performed ODMR scans over the magnetic-field range of 0–60 mT using the ≈ 720 MHz drive frequency for the 8 μm delay line and ≈ 900 MHz drive frequency for the 6 μm delay line. However, measurements performed under identical conditions gave different results, and no repeatable magnetic-field-dependent response could be obtained. We also divided the magnetic-field range into smaller regions, changed the integration time, temperature, magnetic-field angle, and IDT wavelength, and repeated the measurements. Although some signals were observed near the calculated transition positions, most of the measurements were dominated by noise and were not repeatable.

We also performed an MW-excited frequency scan at a fixed in-plane magnetic field of $B = 5.0243$ mT. In this measurement, some distinct features overlapped with the calculated ground-state transitions, including the expected $V2$ ($2 \rightarrow 3$) transition with $\Delta m = \pm 1$. Nevertheless, under these conditions, the features were MW-induced, and the peaks were not sufficient to confirm reliable spin manipulation.

Overall, we successfully fabricated and characterised SAW devices on ZnO/6H-SiC and confirmed the generation, propagation, and focusing of SAWs. We also measured the optical and zero-field responses of the silicon-vacancy V_{Si} centres and performed ODMR measurements at room and cryogenic temperatures. However, under our experimental conditions, a repeatable SAW-driven spin transition could not be demonstrated. In our configuration, the applied magnetic field reduces either the spin coherence time or the luminescence contrast between spin states to the extent that reliable ODMR measurements cannot be obtained.

For future measurements, testing different magnetic-field configurations, such as magnetic field parallel to the c -axis of the substrate, may help to reduce spin-state mixing and obtain a more distinguishable ODMR response. These improvements may allow the SAW-driven transitions predicted by the theoretical calculations to be observed more reliably.

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Appendix A

Supplementary Material

Appendix A.0

Spin-3/2 Hamiltonian Derivation

We consider a spin-3/2 system. The spin quantum number is

$$s = \frac{3}{2}.$$

The allowed magnetic quantum numbers are

$$m = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}.$$

Therefore, the Hilbert space is four-dimensional. We use the ordered basis

$$\left\{ \left| \frac{3}{2} \right\rangle, \left| \frac{1}{2} \right\rangle, \left| -\frac{1}{2} \right\rangle, \left| -\frac{3}{2} \right\rangle \right\}.$$

Throughout the derivation, this ordering is used for all matrix representations.

The spin operators satisfy the angular momentum commutation relations

$$[S_i, S_j] = i\hbar \varepsilon_{ijk} S_k,$$

and

$$\mathbf{S}^2 = S_x^2 + S_y^2 + S_z^2.$$

The basis states are simultaneous eigenstates of $\mathbf{S}^2$ and S_z :

$$\mathbf{S}^2|s, m\rangle = \hbar^2 s(s+1)|s, m\rangle,$$

$$S_z|s, m\rangle = \hbar m|s, m\rangle.$$

For $s = 3/2$,

$$s(s+1) = \frac{3}{2} \left(\frac{3}{2} + 1 \right) = \frac{15}{4}.$$

Thus,

$$\mathbf{S}^2|s, m\rangle = \frac{15}{4}\hbar^2|s, m\rangle.$$

2. Construction of the S_z matrix

The operator S_z is defined through

$$S_z|s, m\rangle = \hbar m|s, m\rangle.$$

This means each basis state $|s, m\rangle$ is an eigenvector of S_z , with eigenvalue $\hbar m$. Using the ordered basis defined above, the diagonal entries are

$$\hbar\frac{3}{2}, \quad \hbar\frac{1}{2}, \quad -\hbar\frac{1}{2}, \quad -\hbar\frac{3}{2}.$$

Therefore,

$$S_z = \hbar \begin{bmatrix} \frac{3}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & -\frac{3}{2} \end{bmatrix}.$$

3. Ladder operators

The ladder operators are defined as

$$S_+ = S_x + iS_y,$$

$$S_- = S_x - iS_y.$$

Their action on the spin basis states is

$$S_{\pm}|s, m\rangle = \hbar\sqrt{s(s+1) - m(m \pm 1)} |s, m \pm 1\rangle.$$

The raising operator S_+ increases m by one, while the lowering operator S_- decreases m by one. For the spin-3/2 system considered here, we use

$$s(s+1) = \frac{15}{4}.$$

4. Explicit action of S_+

The raising operator acts as

$$S_+|s, m\rangle = \hbar\sqrt{s(s+1) - m(m+1)} |s, m+1\rangle.$$

Substituting $s(s+1) = 15/4$ for the four allowed values of m gives

$$S_+ \left| \frac{3}{2} \right\rangle = 0,$$

$$S_+ \left| \frac{1}{2} \right\rangle = \hbar\sqrt{3} \left| \frac{3}{2} \right\rangle,$$

$$S_+ \left| -\frac{1}{2} \right\rangle = 2\hbar \left| \frac{1}{2} \right\rangle,$$

$$S_+ \left| -\frac{3}{2} \right\rangle = \hbar\sqrt{3} \left| -\frac{1}{2} \right\rangle.$$

The first result is zero because there is no state with $m = 5/2$. The other coefficients follow directly from the square-root factor in the ladder-operator formula.

5. Matrix form of S_+

The matrix element in row i , column j represents the amplitude for the operator to take basis state j into basis state i . Using the results above, the first column is zero because $S_+ |3/2\rangle = 0$. The remaining nonzero entries connect neighbouring m -states with coefficients $\hbar\sqrt{3}$, $2\hbar$, and $\hbar\sqrt{3}$. Thus,

$$S_+ = \hbar \begin{bmatrix} 0 & \sqrt{3} & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & \sqrt{3} \\ 0 & 0 & 0 & 0 \end{bmatrix}.$$

6. Explicit action of S_-

The lowering operator acts as

$$S_-|s, m\rangle = \hbar\sqrt{s(s+1) - m(m-1)}|s, m-1\rangle.$$

Again using $s(s+1) = 15/4$, the four actions are

$$S_- \left| \frac{3}{2} \right\rangle = \hbar\sqrt{3} \left| \frac{1}{2} \right\rangle,$$

$$S_- \left| \frac{1}{2} \right\rangle = 2\hbar \left| -\frac{1}{2} \right\rangle,$$

$$S_- \left| -\frac{1}{2} \right\rangle = \hbar\sqrt{3} \left| -\frac{3}{2} \right\rangle,$$

$$S_- \left| -\frac{3}{2} \right\rangle = 0.$$

The final result is zero because there is no state with $m = -5/2$.

7. Matrix form of S_-

Using the same ordered basis, the nonzero entries now appear below the diagonal because S_- lowers the magnetic quantum number. Therefore,

$$S_- = \hbar \begin{bmatrix} 0 & 0 & 0 & 0 \\ \sqrt{3} & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & \sqrt{3} & 0 \end{bmatrix}.$$

This also satisfies

$$S_- = S_+^\dagger.$$

8. Construction of S_x and S_y

From the definitions

$$S_+ = S_x + iS_y,$$

$$S_- = S_x - iS_y,$$

we can solve for S_x and S_y . Adding the equations gives

$$S_x = \frac{S_+ + S_-}{2},$$

while subtracting them gives

$$S_y = \frac{S_+ - S_-}{2i}.$$

Therefore,

$$S_x = \hbar \begin{bmatrix} 0 & \frac{\sqrt{3}}{2} & 0 & 0 \\ \frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & \frac{\sqrt{3}}{2} \\ 0 & 0 & \frac{\sqrt{3}}{2} & 0 \end{bmatrix},$$

and

$$S_y = \hbar \begin{bmatrix} 0 & -\frac{i\sqrt{3}}{2} & 0 & 0 \\ \frac{i\sqrt{3}}{2} & 0 & -i & 0 \\ 0 & i & 0 & -\frac{i\sqrt{3}}{2} \\ 0 & 0 & \frac{i\sqrt{3}}{2} & 0 \end{bmatrix}.$$

9. Dimensionless spin matrices

In many spin-defect calculations, including SiC spin-defect Hamiltonians, it is common to use dimensionless spin matrices. These are defined by

$$S_i^{(d)} = \frac{S_i}{\hbar}.$$

In this convention,

$$S_z^{(d)} = \begin{bmatrix} \frac{3}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & -\frac{3}{2} \end{bmatrix},$$

$$S_x^{(d)} = \begin{bmatrix} 0 & \frac{\sqrt{3}}{2} & 0 & 0 \\ \frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & \frac{\sqrt{3}}{2} \\ 0 & 0 & \frac{\sqrt{3}}{2} & 0 \end{bmatrix},$$

and

$$S_y^{(d)} = \begin{bmatrix} 0 & -\frac{i\sqrt{3}}{2} & 0 & 0 \\ \frac{i\sqrt{3}}{2} & 0 & -i & 0 \\ 0 & i & 0 & -\frac{i\sqrt{3}}{2} \\ 0 & 0 & \frac{i\sqrt{3}}{2} & 0 \end{bmatrix}.$$

For simplicity, from this point onward we use dimensionless spin matrices and write

$$S_x, \quad S_y, \quad S_z$$

instead of

$$S_x^{(d)}, \quad S_y^{(d)}, \quad S_z^{(d)}.$$

This is equivalent to setting

$$\hbar = 1.$$

10. Zero-field splitting term

For a spin- S system with axial zero-field splitting, the standard form of the zero-field splitting Hamiltonian is

$$H_{\text{ZFS}} = D \left(S_z^2 - \frac{S(S+1)}{3} I \right).$$

The origin of the factor $1/3$ can be understood from the total spin relation

$$S_x^2 + S_y^2 + S_z^2 = S(S + 1)I.$$

If the spin contribution is averaged equally over the three spatial directions x , y , and z , the average contribution of one direction is

$$\frac{S_x^2 + S_y^2 + S_z^2}{3} = \frac{S(S + 1)}{3}I.$$

Therefore, the zero-field splitting term subtracts this isotropic average contribution and keeps only the axial anisotropic part along the defect symmetry axis. For $S = 3/2$,

$$S(S + 1) = \frac{3}{2} \frac{5}{2} = \frac{15}{4},$$

and therefore

$$\frac{S(S + 1)}{3} = \frac{5}{4}.$$

Thus,

$$H_{\text{ZFS}} = D \left(S_z^2 - \frac{5}{4}I \right).$$

Using the dimensionless S_z matrix,

$$S_z = \begin{bmatrix} \frac{3}{2} & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & -\frac{3}{2} \end{bmatrix},$$

we obtain

$$S_z^2 = \begin{bmatrix} \frac{9}{4} & 0 & 0 & 0 \\ 0 & \frac{1}{4} & 0 & 0 \\ 0 & 0 & \frac{1}{4} & 0 \\ 0 & 0 & 0 & \frac{9}{4} \end{bmatrix}.$$

Subtracting $\frac{5}{4}I$ gives

$$S_z^2 - \frac{5}{4}I = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} = \text{diag}(1, -1, -1, 1).$$

Thus, the zero-field splitting Hamiltonian is

$$H_{\text{ZFS}} = D \text{diag}(1, -1, -1, 1).$$

This gives the zero-field energies

$$E_{\pm 3/2} = D,$$

and

$$E_{\pm 1/2} = -D.$$

Therefore, the energy separation between the two doublets is

$$\Delta E = D - (-D) = 2D.$$

11. Zeeman Hamiltonian

The Zeeman interaction is

$$H_Z = g\mu_B \mathbf{B} \cdot \mathbf{S}.$$

For a magnetic field

$$\mathbf{B} = (B_x, B_y, B_z),$$

we have

$$\mathbf{B} \cdot \mathbf{S} = B_x S_x + B_y S_y + B_z S_z.$$

Therefore,

$$H_Z = g\mu_B (B_x S_x + B_y S_y + B_z S_z).$$

12. Full Hamiltonian

The full spin-3/2 Hamiltonian is the sum of the Zeeman term and the zero-field splitting term:

$$H = H_Z + H_{\text{ZFS}}.$$

Therefore,

$$H = g\mu_B (B_x S_x + B_y S_y + B_z S_z) + D \left(S_z^2 - \frac{S(S+1)}{3} I \right).$$

For $S = 3/2$,

$$H = g\mu_B (B_x S_x + B_y S_y + B_z S_z) + D \left(S_z^2 - \frac{5}{4} I \right).$$

This is the standard axial spin-3/2 Hamiltonian used for SiC spin defects.

13. Explicit matrix form of the full Hamiltonian

Using the dimensionless spin matrices derived above, we now compute

$$g\mu_B (B_x S_x + B_y S_y + B_z S_z).$$

The $B_x S_x + B_y S_y$ part gives the off-diagonal terms

$$\frac{\sqrt{3}}{2} g\mu_B (B_x - iB_y), \quad g\mu_B (B_x - iB_y), \quad \frac{\sqrt{3}}{2} g\mu_B (B_x - iB_y),$$

above the diagonal, and their complex conjugates below the diagonal. The $B_z S_z$ part gives the diagonal terms

$$\frac{3}{2} g\mu_B B_z, \quad \frac{1}{2} g\mu_B B_z, \quad -\frac{1}{2} g\mu_B B_z, \quad -\frac{3}{2} g\mu_B B_z.$$

The zero-field splitting part gives

$$D \text{diag}(1, -1, -1, 1).$$

Thus, the full Hamiltonian matrix is

$$H = \begin{bmatrix} D + \frac{3}{2}g\mu_B B_z & \frac{\sqrt{3}}{2}g\mu_B(B_x - iB_y) & 0 & 0 \\ \frac{\sqrt{3}}{2}g\mu_B(B_x + iB_y) & -D + \frac{1}{2}g\mu_B B_z & g\mu_B(B_x - iB_y) & 0 \\ 0 & g\mu_B(B_x + iB_y) & -D - \frac{1}{2}g\mu_B B_z & \frac{\sqrt{3}}{2}g\mu_B(B_x - iB_y) \\ 0 & 0 & \frac{\sqrt{3}}{2}g\mu_B(B_x + iB_y) & D - \frac{3}{2}g\mu_B B_z \end{bmatrix}.$$

14. Special case: magnetic field along the x -axis

Let

$$\mathbf{B} = (B_x, 0, 0).$$

Then $B_y = 0$ and $B_z = 0$. We define

$$\alpha = g\mu_B B_x.$$

The Hamiltonian becomes

$$H = \alpha S_x + D \left(S_z^2 - \frac{5}{4}I \right).$$

Using

$$S_z^2 - \frac{5}{4}I = \text{diag}(1, -1, -1, 1),$$

we get

$$H = \alpha \begin{bmatrix} 0 & \frac{\sqrt{3}}{2} & 0 & 0 \\ \frac{\sqrt{3}}{2} & 0 & 1 & 0 \\ 0 & 1 & 0 & \frac{\sqrt{3}}{2} \\ 0 & 0 & \frac{\sqrt{3}}{2} & 0 \end{bmatrix} + D \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}.$$

Therefore,

$$H = \begin{bmatrix} D & \frac{\sqrt{3}}{2}\alpha & 0 & 0 \\ \frac{\sqrt{3}}{2}\alpha & -D & \alpha & 0 \\ 0 & \alpha & -D & \frac{\sqrt{3}}{2}\alpha \\ 0 & 0 & \frac{\sqrt{3}}{2}\alpha & D \end{bmatrix}.$$

15. Block diagonalization for magnetic field along x

For a magnetic field along x , the Hamiltonian is symmetric under the exchange

$$m \rightarrow -m.$$

Therefore, we can use symmetric and antisymmetric combinations of states. Define

$$\begin{aligned} |\psi_1^+\rangle &= \frac{|\frac{3}{2}\rangle + |-\frac{3}{2}\rangle}{\sqrt{2}}, & |\psi_2^+\rangle &= \frac{|\frac{1}{2}\rangle + |-\frac{1}{2}\rangle}{\sqrt{2}}, \\ |\psi_1^-\rangle &= \frac{|\frac{3}{2}\rangle - |-\frac{3}{2}\rangle}{\sqrt{2}}, & |\psi_2^-\rangle &= \frac{|\frac{1}{2}\rangle - |-\frac{1}{2}\rangle}{\sqrt{2}}. \end{aligned}$$

In the ordered parity basis

$$\{|\psi_1^+\rangle, |\psi_2^+\rangle, |\psi_1^-\rangle, |\psi_2^-\rangle\},$$

the Hamiltonian becomes block diagonal:

$$H = \begin{bmatrix} D & \frac{\sqrt{3}}{2}\alpha & 0 & 0 \\ \frac{\sqrt{3}}{2}\alpha & -D + \alpha & 0 & 0 \\ 0 & 0 & D & \frac{\sqrt{3}}{2}\alpha \\ 0 & 0 & \frac{\sqrt{3}}{2}\alpha & -D - \alpha \end{bmatrix}.$$

Equivalently,

$$H = \begin{bmatrix} \boxed{\begin{matrix} D & \frac{\sqrt{3}}{2}\alpha \\ \frac{\sqrt{3}}{2}\alpha & -D + \alpha \end{matrix}} & 0 \\ 0 & \boxed{\begin{matrix} D & \frac{\sqrt{3}}{2}\alpha \\ \frac{\sqrt{3}}{2}\alpha & -D - \alpha \end{matrix}} \end{bmatrix}.$$

16. Eigenvalues for magnetic field along x

The positive-parity block is

$$H_+ = \begin{bmatrix} D & \frac{\sqrt{3}}{2}\alpha \\ \frac{\sqrt{3}}{2}\alpha & -D + \alpha \end{bmatrix}.$$

For a general 2×2 matrix

$$\begin{bmatrix} a & c \\ c & b \end{bmatrix},$$

the eigenvalues are

$$E_{\pm} = \frac{a+b}{2} \pm \sqrt{\left(\frac{a-b}{2}\right)^2 + c^2}.$$

For H_+ , we identify

$$a = D, \quad b = -D + \alpha, \quad c = \frac{\sqrt{3}}{2}\alpha.$$

Thus,

$$\frac{a+b}{2} = \frac{\alpha}{2}, \quad \frac{a-b}{2} = D - \frac{\alpha}{2}.$$

Therefore,

$$E_{\pm}^{(+)} = \frac{\alpha}{2} \pm \sqrt{\left(D - \frac{\alpha}{2}\right)^2 + \frac{3}{4}\alpha^2}.$$

Simplifying the square root gives

$$\left(D - \frac{\alpha}{2}\right)^2 + \frac{3}{4}\alpha^2 = D^2 - D\alpha + \alpha^2.$$

Hence,

$$E_{\pm}^{(+)} = \frac{\alpha}{2} \pm \sqrt{D^2 - D\alpha + \alpha^2}.$$

The negative-parity block is

$$H_- = \begin{bmatrix} D & \frac{\sqrt{3}}{2}\alpha \\ \frac{\sqrt{3}}{2}\alpha & -D - \alpha \end{bmatrix}.$$

For H_- , we identify

$$a = D, \quad b = -D - \alpha, \quad c = \frac{\sqrt{3}}{2}\alpha.$$

Thus,

$$\frac{a+b}{2} = -\frac{\alpha}{2}, \quad \frac{a-b}{2} = D + \frac{\alpha}{2}.$$

Therefore,

$$E_{\pm}^{(-)} = -\frac{\alpha}{2} \pm \sqrt{\left(D + \frac{\alpha}{2}\right)^2 + \frac{3}{4}\alpha^2}.$$

Simplifying the square root gives

$$\left(D + \frac{\alpha}{2}\right)^2 + \frac{3}{4}\alpha^2 = D^2 + D\alpha + \alpha^2.$$

Hence,

$$E_{\pm}^{(-)} = -\frac{\alpha}{2} \pm \sqrt{D^2 + D\alpha + \alpha^2}.$$

Therefore, the four eigenvalues for a magnetic field along x are

$$E_{\pm}^{(+)} = \frac{\alpha}{2} \pm \sqrt{D^2 - D\alpha + \alpha^2},$$

and

$$E_{\pm}^{(-)} = -\frac{\alpha}{2} \pm \sqrt{D^2 + D\alpha + \alpha^2}.$$

17. Final Hamiltonian

The final spin-3/2 Hamiltonian is

$$H = g\mu_B (B_x S_x + B_y S_y + B_z S_z) + D \left(S_z^2 - \frac{S(S+1)}{3} I \right) \quad (\text{A.1})$$

with

$$S = \frac{3}{2}.$$

Therefore,

$$H = g\mu_B (B_x S_x + B_y S_y + B_z S_z) + D \left(S_z^2 - \frac{5}{4} I \right)$$

or, equivalently,

$$H = g\mu_B \mathbf{B} \cdot \mathbf{S} + D \left(S_z^2 - \frac{5}{4} I \right).$$

Appendix B

Graphs

B.1 V2 GS

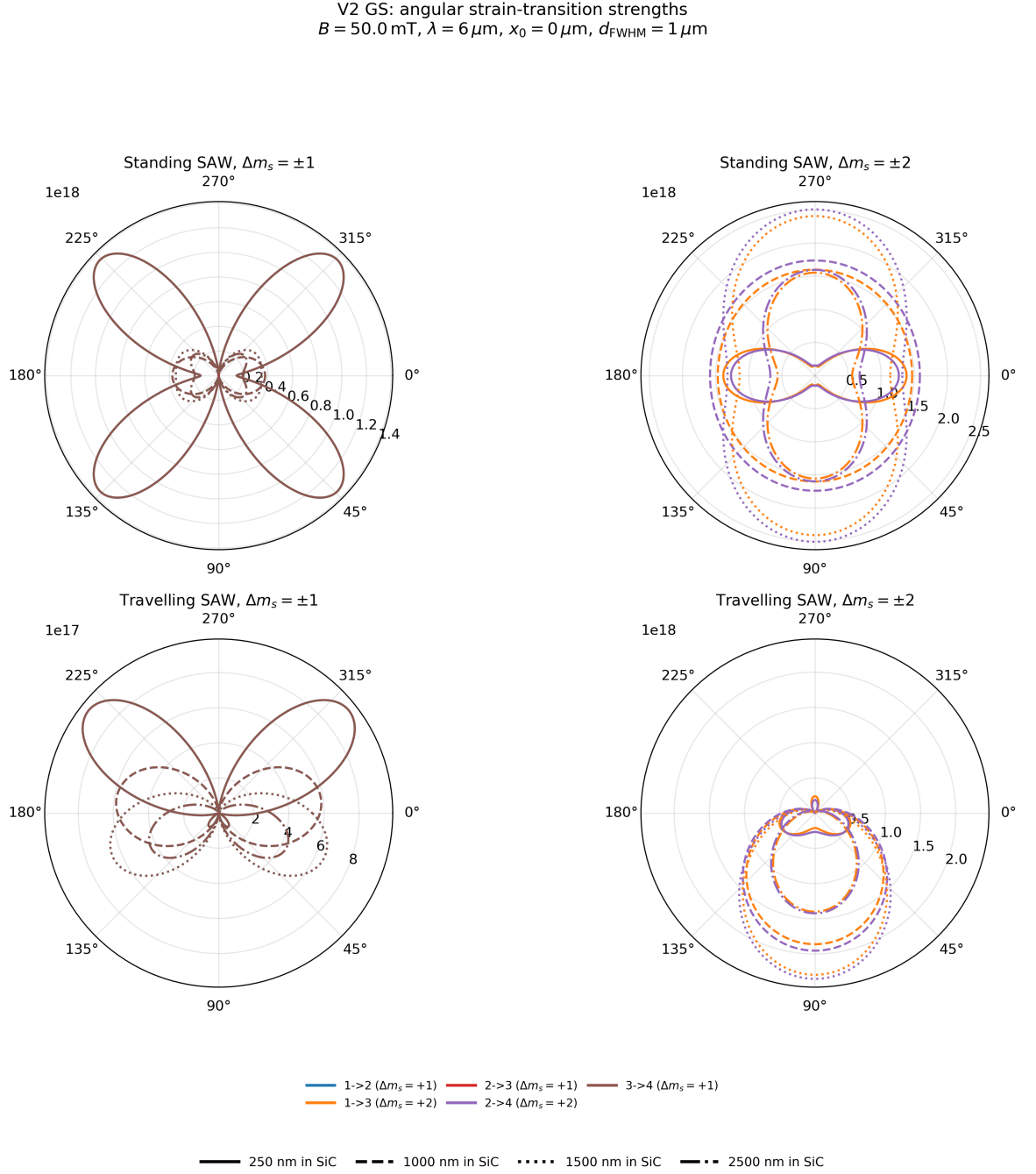


Figure B.1: Angular dependence of the SAW-driven transition strengths for the V2 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

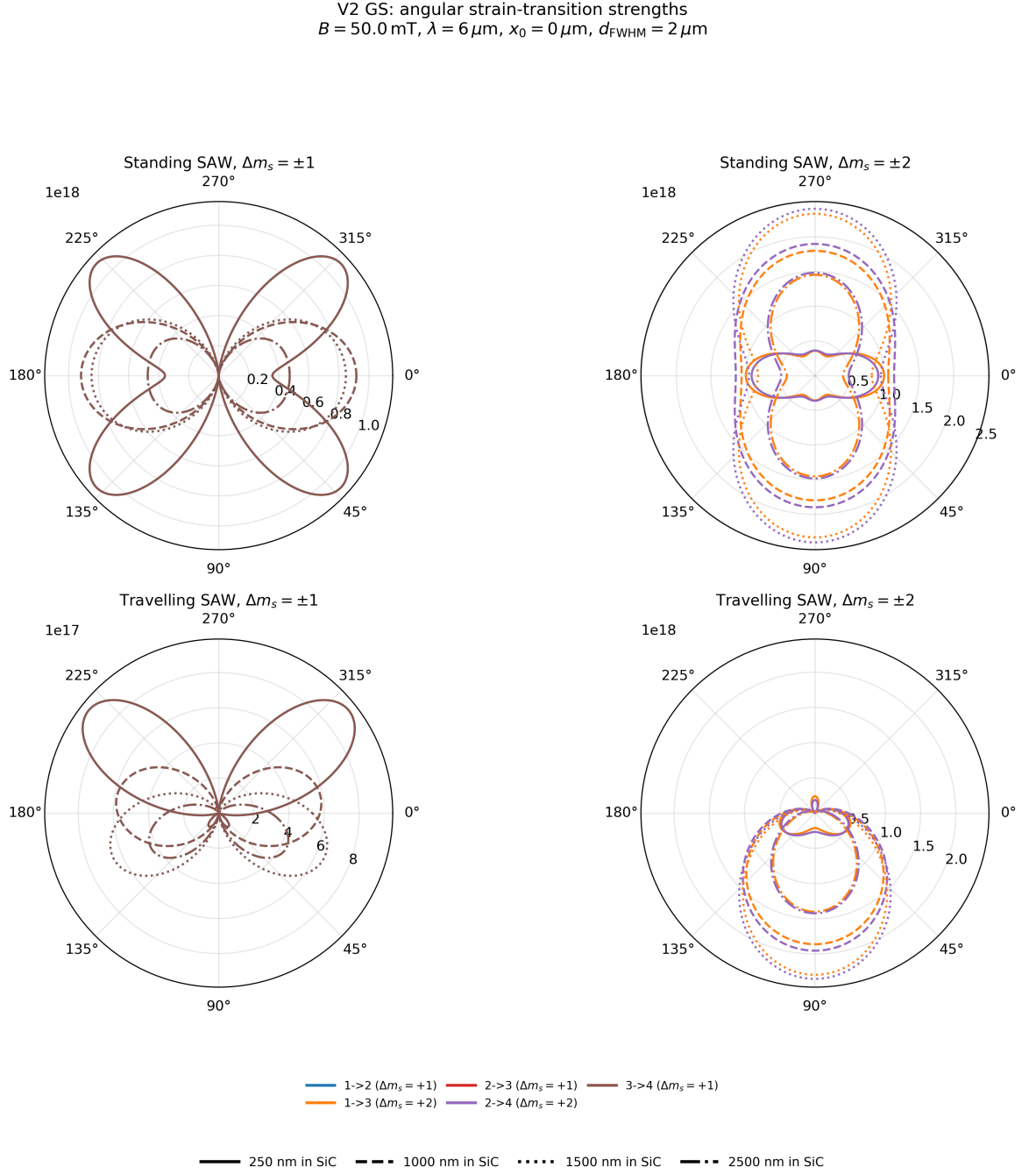


Figure B.2: Angular dependence of the SAW-driven transition strengths for the V2 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $2 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

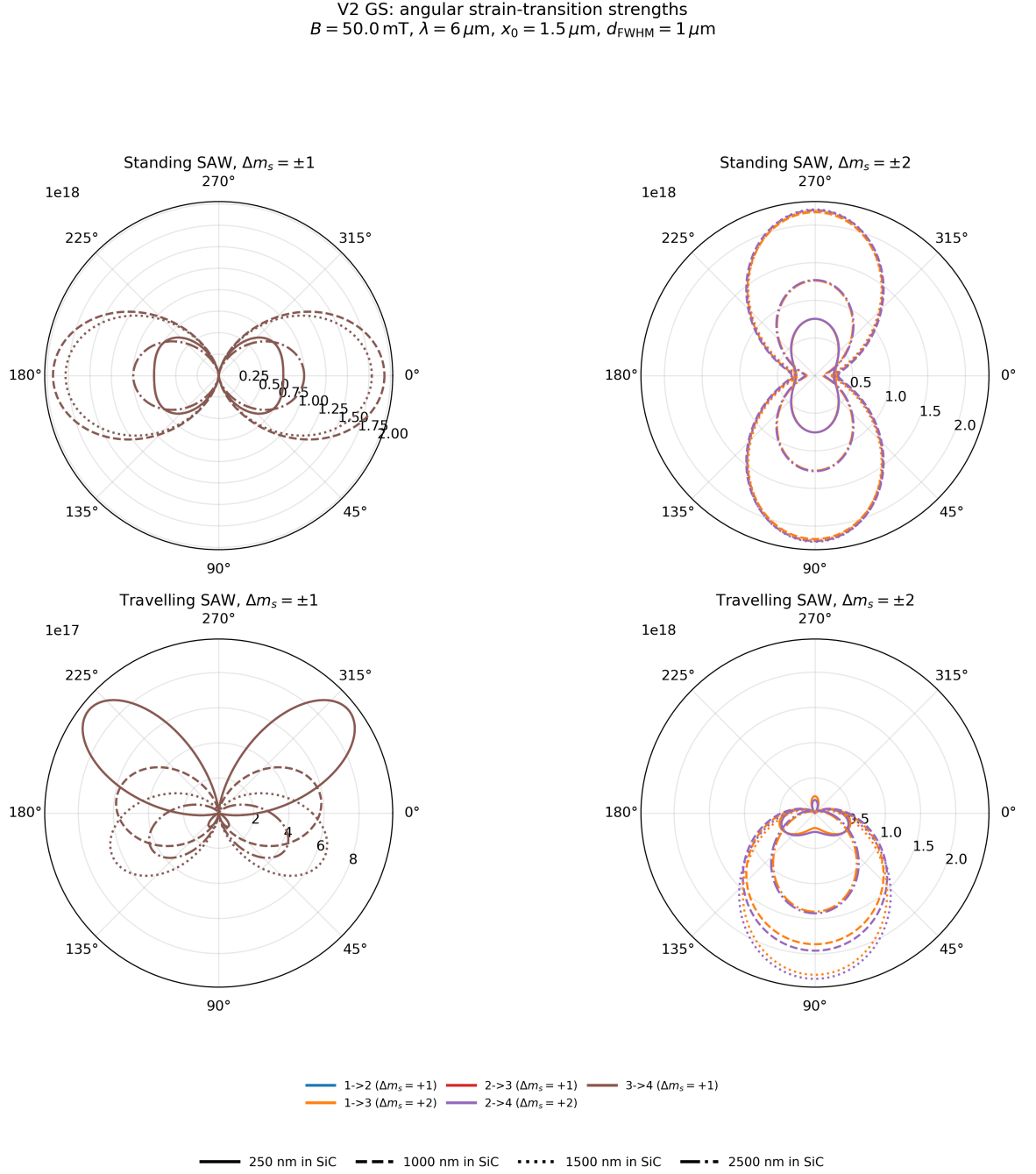


Figure B.3: Angular dependence of the SAW-driven transition strengths for the V2 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 1.5 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

B.2 V2 ES

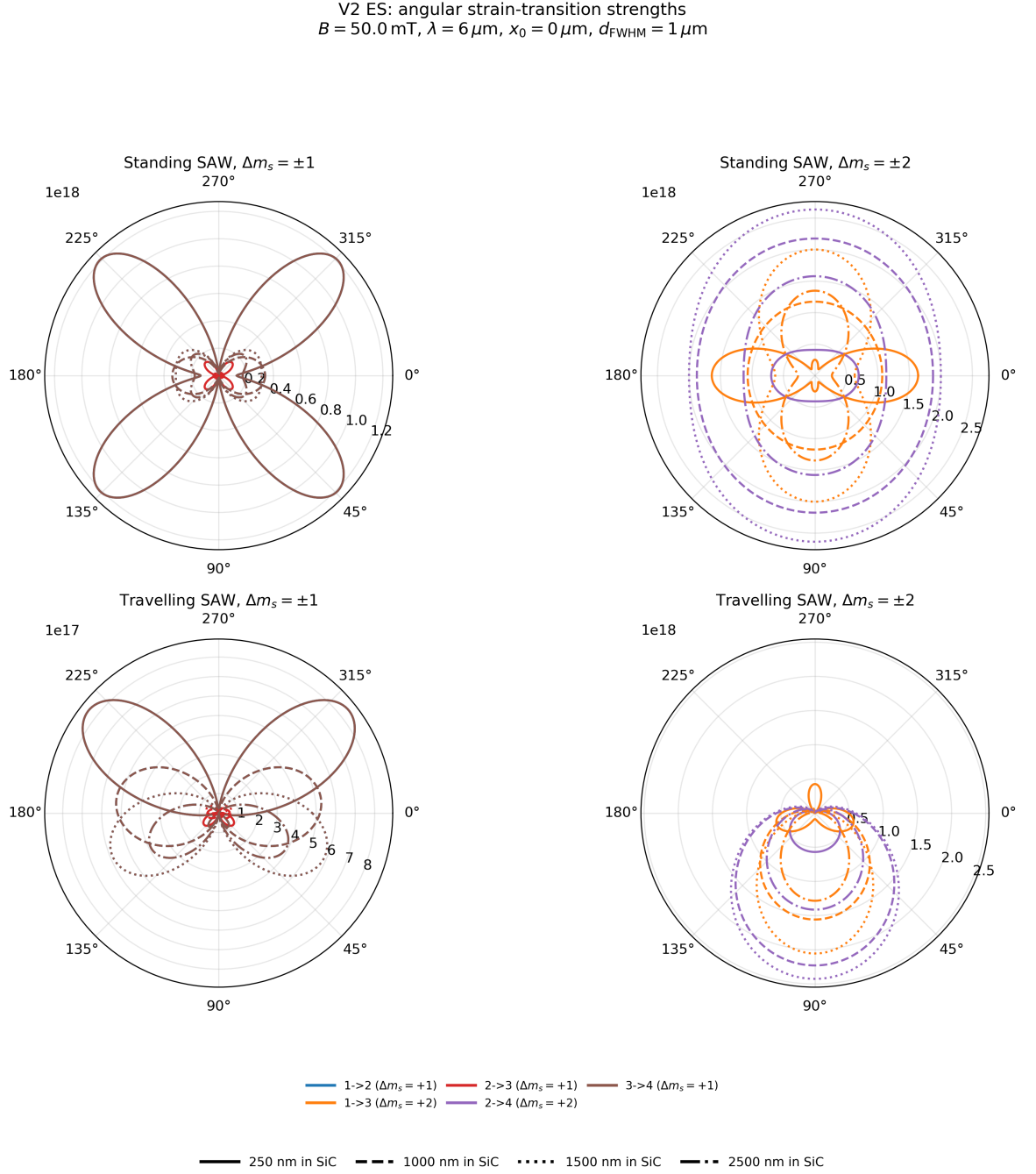


Figure B.4: Angular dependence of the SAW-driven transition strengths for the V2 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

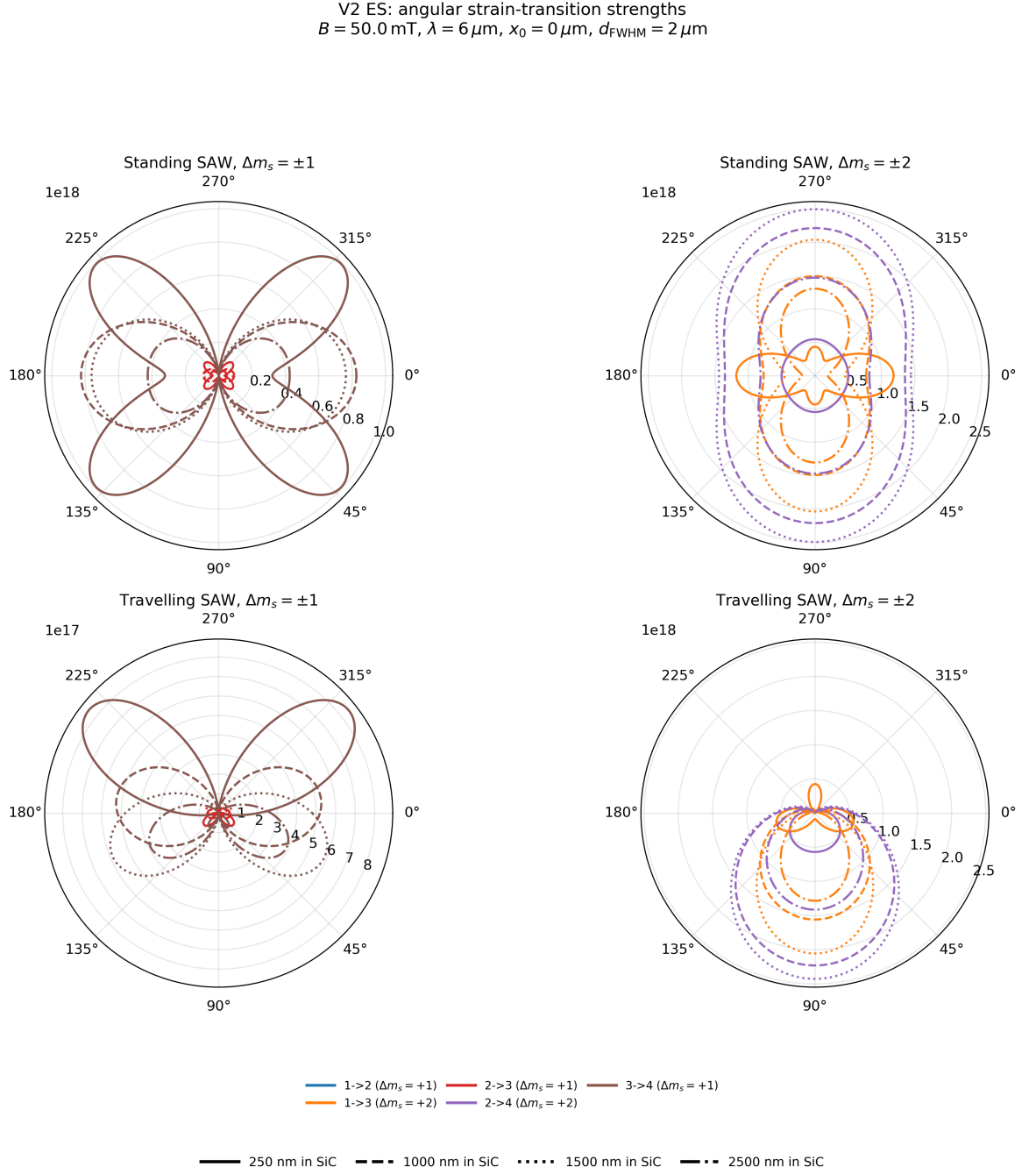


Figure B.5: Angular dependence of the SAW-driven transition strengths for the V2 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $2 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

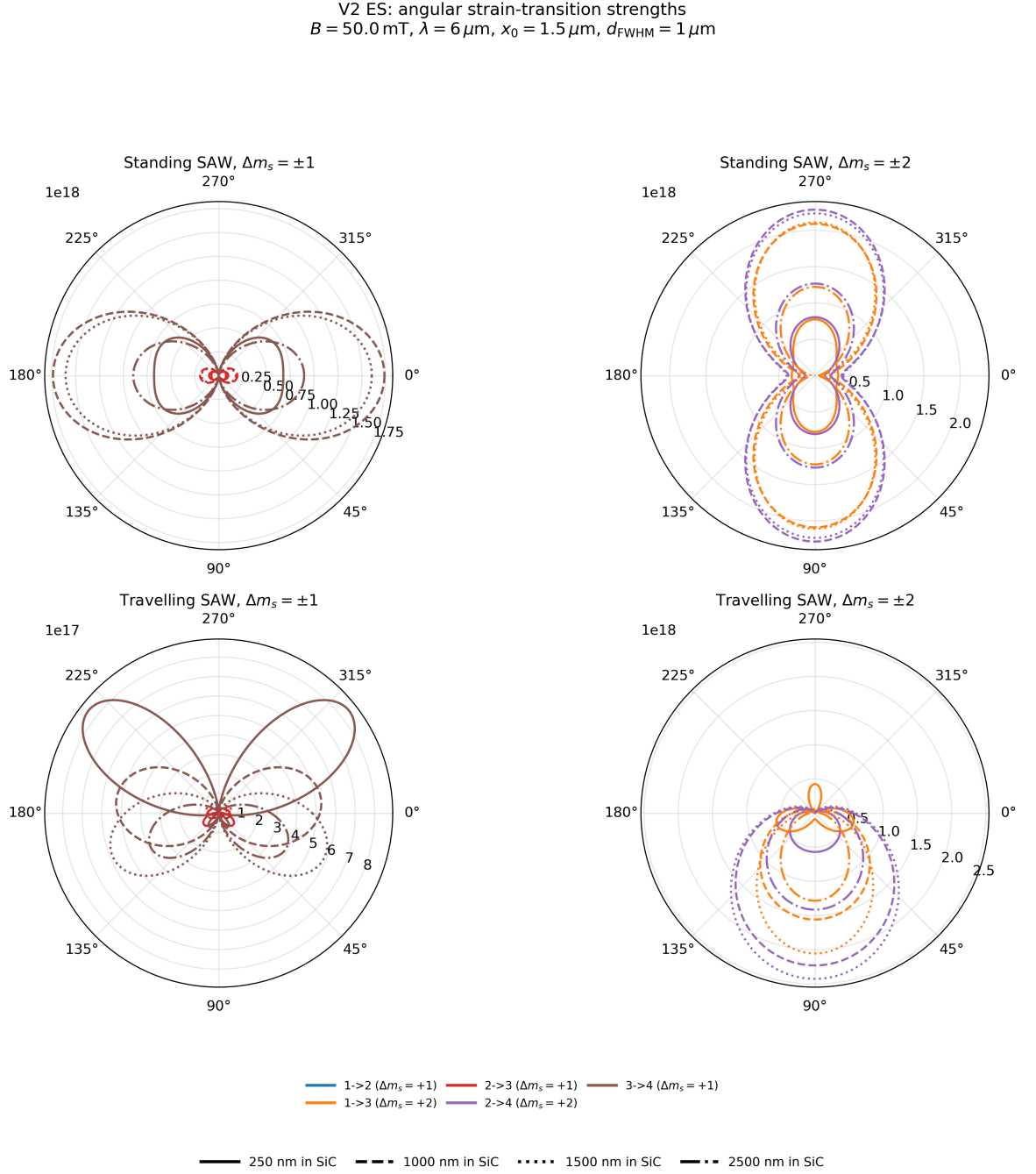


Figure B.6: Angular dependence of the SAW-driven transition strengths for the V2 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 1.5 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

B.3 V3 GS

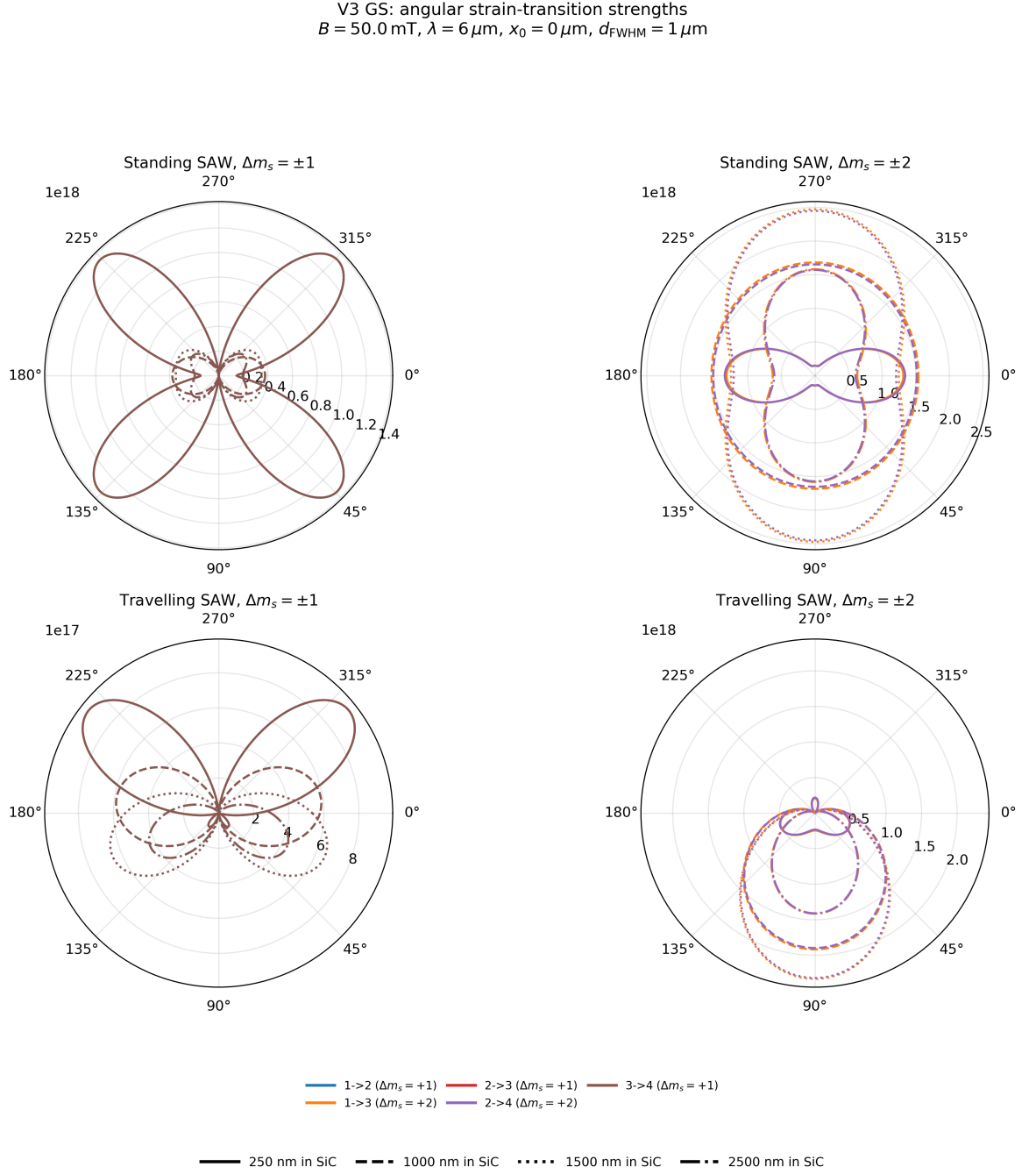


Figure B.7: Angular dependence of the SAW-driven transition strengths for the V3 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

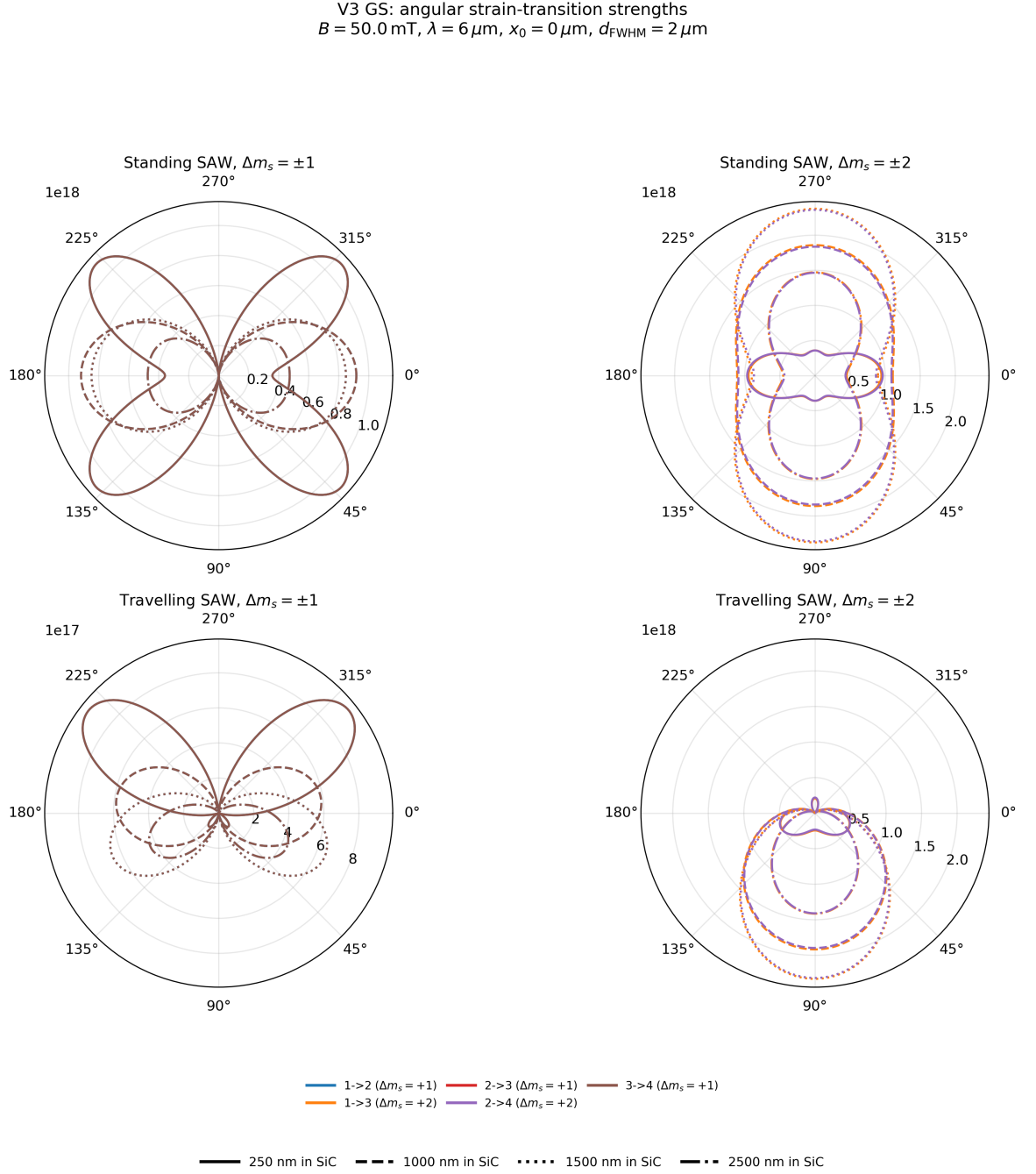


Figure B.8: Angular dependence of the SAW-driven transition strengths for the V3 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $2 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

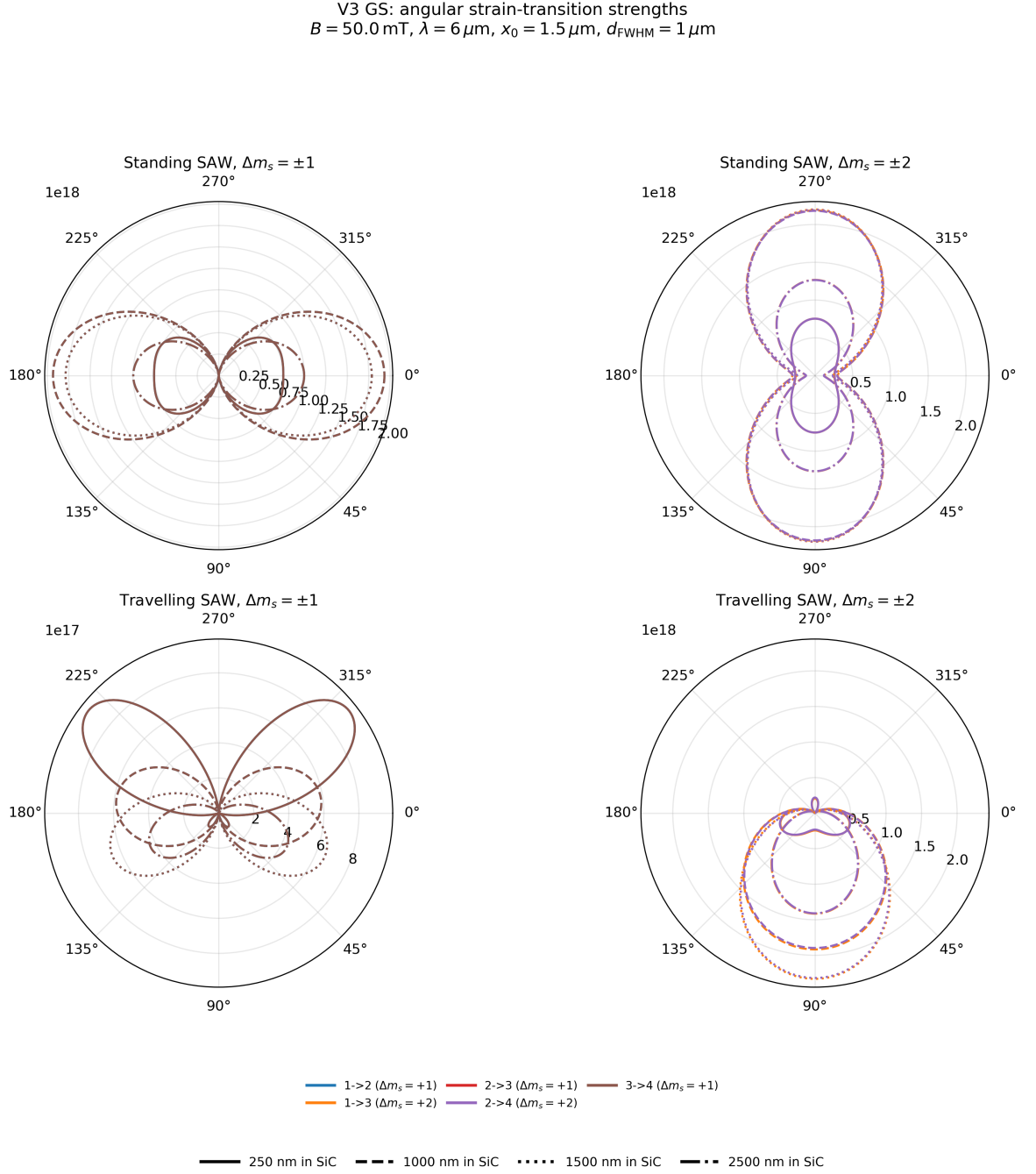


Figure B.9: Angular dependence of the SAW-driven transition strengths for the V3 centre in the ground state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 1.5 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

B.4 V3 ES

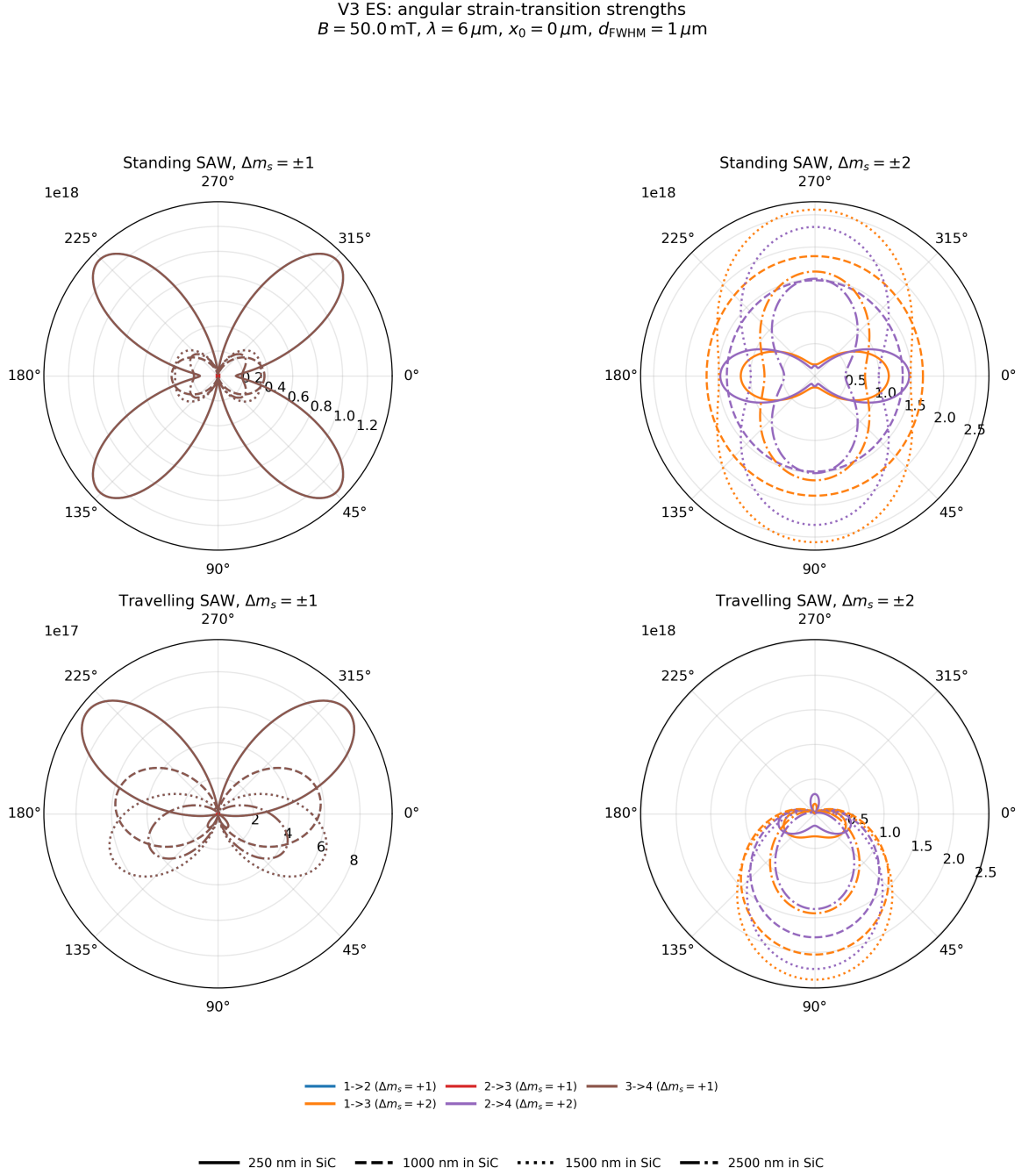


Figure B.10: Angular dependence of the SAW-driven transition strengths for the V3 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

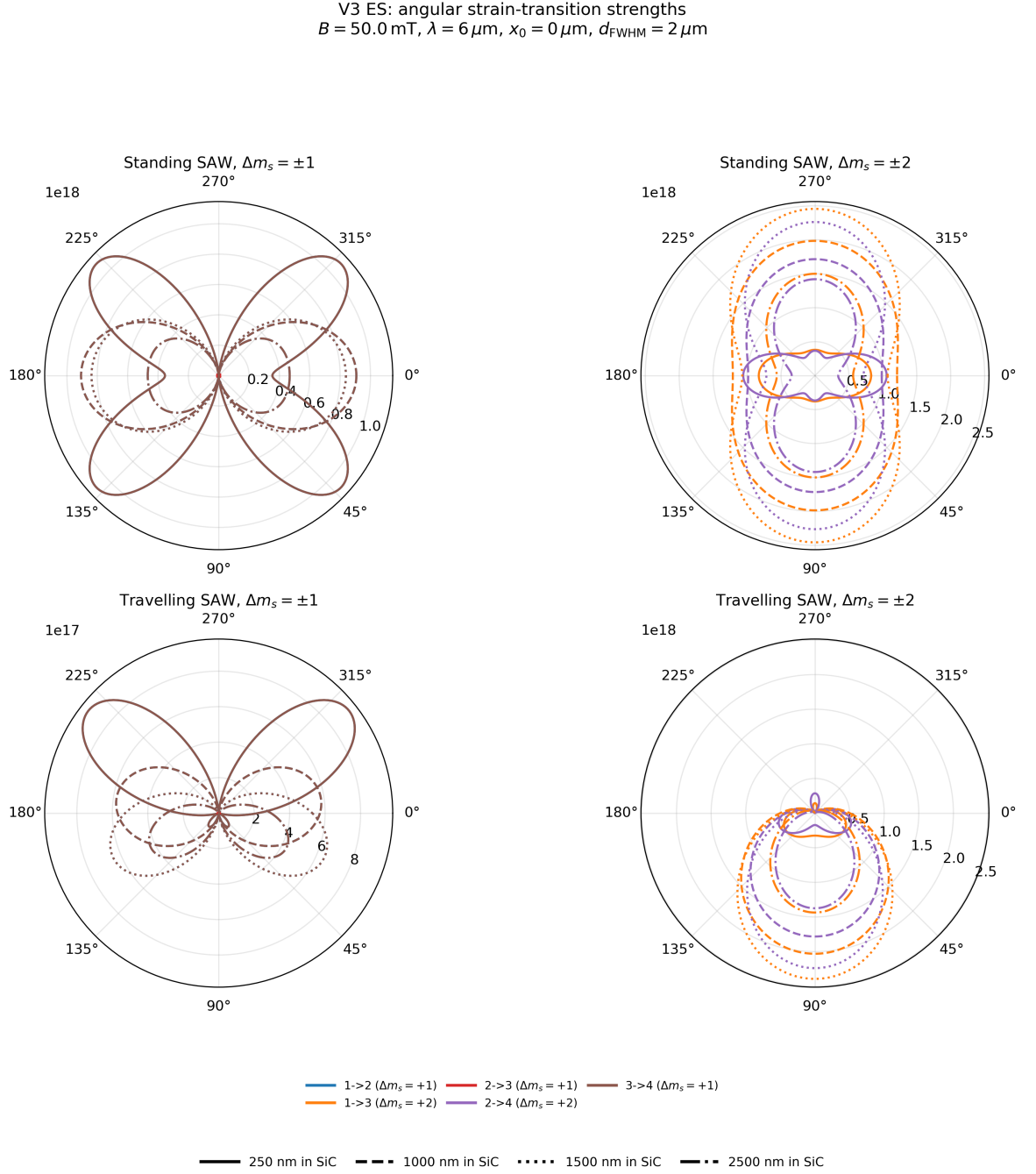


Figure B.11: Angular dependence of the SAW-driven transition strengths for the V3 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 0 \mu\text{m}$ and an intensity FWHM of $2 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

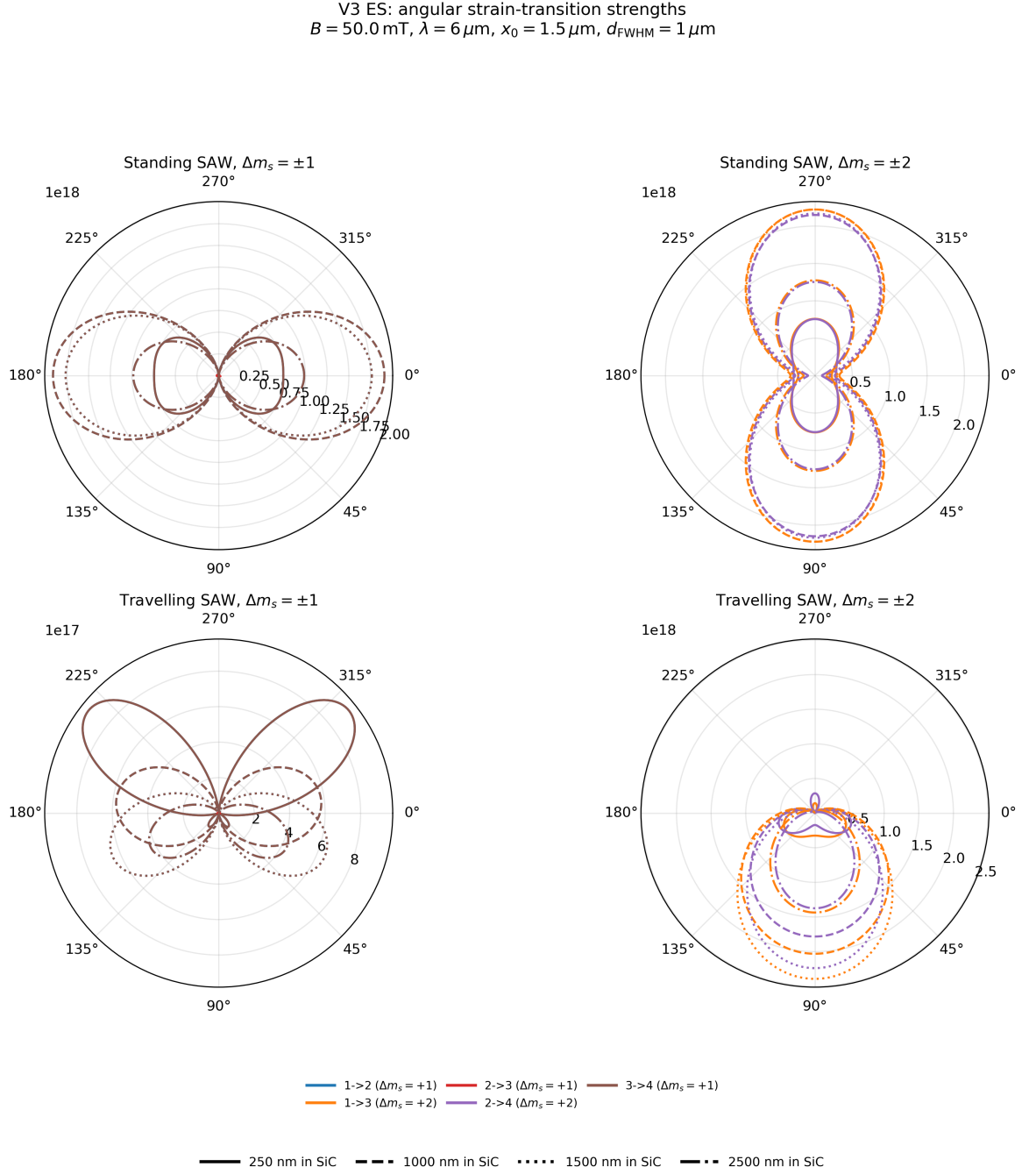


Figure B.12: Angular dependence of the SAW-driven transition strengths for the V3 centre in the excited state at $B = 50 \text{ mT}$, with the laser centred at $x_0 = 1.5 \mu\text{m}$ and an intensity FWHM of $1 \mu\text{m}$. The panels compare standing and travelling SAWs for nominal $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions at different depths inside SiC.

Appendix C

Mathematical and Computational Procedure for Spatially Averaged SAW-Driven Spin Transitions

C.1 Purpose of the Calculations

In this chapter we will be demonstrating how the computational procedure was conducted for the calculations of the angular dependence of the SAW-driven transition strengths shown in Fig. 3.6.

The purpose of the calculations are to determine how the strength of strain-driven spin transitions changes as the direction of an in-plane magnetic field is rotated.

The calculations are conducted for:

- V2 ground state;
- V2 excited state;
- V3 ground state;
- V3 excited state;
- several defect depths inside SiC;
- a standing surface acoustic wave (SAW);
- a travelling SAW.

For both standing and travelling wave configurations, the incoming laser illuminates a finite range of lateral positions. In an ideal scenario where the beam is travelling uniformly, position changes only the common acoustic phase which cancels from the local transition strength. Consequently, normalised incoherent spatial averaging does not change the result. For the standing wave however, finite-spot spatial averaging directly modifies the measured transition strength. Therefore calculations for the standing wave also includes the finite diameter and position of the optical laser spot. The laser does not probe a single point. Instead, it illuminates a range of lateral positions over which the normal and shear strain components change.

For each magnetic-field angle ϕ_B , the strain-driven transition intensity between eigenstates $|i(\phi_B)\rangle$ and $|j(\phi_B)\rangle$ is defined as

$$I_{ij}(\phi_B) = \left| \langle j(\phi_B) | \hat{O} | i(\phi_B) \rangle \right|^2, \quad (\text{C.1})$$

where $\hat{O}$ denotes the relevant strain-driving operator. For a travelling wave, $\hat{O} = \hat{O}_{\text{trav}}$. For a standing wave, the operator depends on position, $\hat{O} = \hat{O}_{\text{stand}}(x)$, and the resulting local intensity is subsequently averaged over the finite Gaussian laser profile.

As $\hat{O}$ is expressed in frequency units, the quantity I_{ij} has units of Hz^2 , representing a relative transition strength, not a normalized experimental intensity.

The depth-dependent strain components are imported from three text-based .out files¹:

$$S_1 = u_{xx}, \quad (\text{C.2})$$

$$S_3 = u_{zz}, \quad (\text{C.3})$$

$$S_5 = 2u_{xz}. \quad (\text{C.4})$$

Assuming that S_5 denotes engineering shear strain, the tensor shear strain used in the spin-strain Hamiltonian is obtained from

$$u_{xz} = \frac{S_5}{2}. \quad (\text{C.5})$$

This conversion is implemented as

```
profile = pd.DataFrame(
    {
        "z_um": df_xx["z_um"],
        "u_xx": df_xx["u_xx"],
        "u_zz": df_zz["u_zz"],
        "u_xz": df_s5["S5"] / 2.0,
    }
)
```

It is important not to divide u_{xz} by two again later in the calculation as it is already divided by the definition.

We have to take account the SiO_2 layer. The depth inside the SiC is calculated from the absolute coordinate using

$$z_{\text{SiC}} = 1000 (z_{\mu\text{m}} - z_{\text{SiO}_2}), \quad (\text{C.6})$$

where the factor 1000 converts micrometres to nanometres.

In code,

```
profile["z_sic_nm"] = (
    profile["z_um"] - SI02_BOTTOM_UM
) * 1000.0
```

¹The data were provided by PD Dr. habil. Georgy Astakhov, Helmholtz- Zentrum Dresden-Rossendorf (HZDR).

The code then selects the available point in the data set which is closest to the requested SiC depth.

C.2 Step 2: Spin Operators

The system is characterised by spin

$$S = \frac{3}{2}. \quad (C.7)$$

The Hilbert-space dimension is

$$2S + 1 = 4. \quad (C.8)$$

The spin matrices are S_x , S_y , and S_z . The calculation also uses

$$S_x^2 = S_x S_x, \quad (C.9)$$

$$S_z^2 = S_z S_z, \quad (C.10)$$

$$S_{xz}^{\text{sym}} = \frac{1}{2} (S_x S_z + S_z S_x). \quad (C.11)$$

They are constructed using QuTiP:

```
SPIN = 3 / 2
DIM = 4

IDENTITY = qeye(DIM)

Sx = jmat(SPIN, "x")
Sy = jmat(SPIN, "y")
Sz = jmat(SPIN, "z")

Sx2 = (Sx * Sx).tidyup()
Sz2 = (Sz * Sz).tidyup()

SxSz_sym = (
    0.5 * (Sx * Sz + Sz * Sx)
).tidyup()
```

C.3 Step 3: Temperature-Dependent ZFS Parameter

The effective zero-field-splitting coefficient is

$$D(T) = D_{\text{ref}} + \beta (T - T_{\text{ref}}) . \quad (\text{C.12})$$

This is implemented as

```
def D_eff_hz(
    center,
    level,
    temperature_K,
    reference_temperature_K=300.0,
):
    key = (center, level)

    return (
        D_REF_HZ[key]
        + BETA_HZ_PER_K[key]
        * (
            temperature_K
            - reference_temperature_K
        )
    )
```

The parameters D_{ref} and β are selected using the pair (center, level).

The zero-field-splitting Hamiltonian is

$$\frac{H_{\text{ZFS}}}{h} = D(T) \left[S_z^2 - \frac{S(S+1)}{3} I \right] . \quad (\text{C.13})$$

For $S = 3/2$, the separation between the $|m_s| = 1/2$ and $|m_s| = 3/2$ doublets is

$$\Delta f_{\text{ZFS}} = 2|D| . \quad (\text{C.14})$$

Thus the numerical values stored in D_REF_HZ must be Hamiltonian D -coefficients.

C.4 Step 4: Magnetic-Field Hamiltonian

The magnetic field is assumed to lie in the xy -plane:

$$B_x = B \cos \phi_B, \quad (\text{C.15})$$

$$B_y = B \sin \phi_B, \quad (\text{C.16})$$

$$B_z = 0. \quad (\text{C.17})$$

The gyromagnetic ratio in frequency units is

$$\gamma = \frac{g\mu_B}{h}. \quad (\text{C.18})$$

The Zeeman Hamiltonian is

$$\frac{H_{\text{Zeeman}}}{h} = \gamma (B_x S_x + B_y S_y + B_z S_z). \quad (\text{C.19})$$

The complete static Hamiltonian is

$$\frac{H_0}{h} = D(T) \left[S_z^2 - \frac{S(S+1)}{3} I \right] + \gamma (B_x S_x + B_y S_y + B_z S_z). \quad (\text{C.20})$$

This is implemented as

```
def static_hamiltonian_over_h(
    B_T,
    phi_B_rad,
    D_hz,
):
    Bx_T = B_T * np.cos(phi_B_rad)
    By_T = B_T * np.sin(phi_B_rad)
    Bz_T = 0.0

    H_zeeman = GYROMAGNETIC_RATIO_HZ_PER_T * (
        Bx_T * Sx
        + By_T * Sy
        + Bz_T * Sz
    )

    H_zfs = D_hz * (
        Sz2
        - (
            SPIN * (SPIN + 1) / 3.0
        ) * IDENTITY
```

```
)

return (H_zfs + H_zeeman).tidyup()
```

C.5 Step 5: Diagonalization and Transition Frequencies

For each magnetic-field angle, the code solves

$$\frac{H_0}{h}|i\rangle = f_i|i\rangle, \quad (\text{C.21})$$

where $f_i = E_i/h$ is expressed in hertz.

In code,

```
eigenvalues, eigenstates = H0.eigenstates()
```

For every pair $j > i$, the transition frequency is

$$f_{ij} = f_j - f_i. \quad (\text{C.22})$$

The implementation is

```
frequency_hz = float(
    np.real(
        energies_hz[j] - energies_hz[i]
    )
)
```

There are six unique pairs for a four-level system:

$$(0, 1), (0, 2), (0, 3), (1, 2), (1, 3), (2, 3). \quad (\text{C.23})$$

C.6 Step 6: Local Spin-Strain Operator

The local spin–strain interaction is

$$\hat{O}_{\text{strain}} = a \left[u_{xx} S_x^2 + u_{zz} S_z^2 + u_{xz} \frac{S_x S_z + S_z S_x}{2} \right]. \quad (\text{C.24})$$

The coupling constant is represented by

$$a = \text{SPIN_PHONON_COUPLING_HZ_PER_STRAIN} \quad (\text{C.25})$$

In the absence of a solid measurement for a spin-phonon coupling constant for 6H-SiC in the literature, $a = 8.5 \times 10^{12}$ Hz strain⁻¹ in terahertz regime, is adopted as an effective phenomenological value, corresponding to a coupling scale of 8.5 MHz at a strain of 10^{-6} .

The implementation is

```
def strain_operator_local(a_hz, strain):
    u_xx, u_zz, u_xz = strain

    return (
        a_hz
        * (
            u_xx * Sx2
            + u_zz * Sz2
            + u_xz * SxSz_sym
        )
    ).tidyup()
```

C.7 Step 7: Travelling-Wave Operator

The normal and shear strains are approximately 90° out of phase.

A travelling-wave representation is therefore

$$u_{xx}(x, t) = u_{xx} \cos(kx - \omega t), \quad (\text{C.26})$$

$$u_{zz}(x, t) = u_{zz} \cos(kx - \omega t), \quad (\text{C.27})$$

$$u_{xz}(x, t) = u_{xz} \sin(kx - \omega t). \quad (\text{C.28})$$

The corresponding complex positive-frequency drive amplitude is written

$$O_+^{\text{trav}} = a \left[u_{xx} S_x^2 + u_{zz} S_z^2 + i u_{xz} \frac{S_x S_z + S_z S_x}{2} \right]. \quad (\text{C.29})$$

This is implemented as

```
def strain_operator_travelling(
    a_hz,
    strain,
):
    u_xx, u_zz, u_xz = strain
```

```

complex_strain = (
    u_xx,
    u_zz,
    1j * u_xz,
)

return strain_operator_local(
    a_hz,
    complex_strain,
)

```

The sign of i depends on the Fourier convention and propagation direction. Reversing the propagation direction may replace $+i$ by $-i$.

The operator $\hat{O}_+^{\text{trav}}$ is not, by itself, the complete Hermitian time-dependent Hamiltonian. The physical drive is

$$\frac{H_{\text{drive}}(t)}{h} = O_+^{\text{trav}} e^{-i\omega t} + (O_+^{\text{trav}})^\dagger e^{+i\omega t}. \quad (\text{C.30})$$

For transition strength calculations, the positive frequency amplitude is used as

$$I_{ij}^{\text{trav}} = |\langle j | O_+^{\text{trav}} | i \rangle|^2. \quad (\text{C.31})$$

For an ideal uniform travelling wave, moving along x introduces a common phase e^{ikx} . Because

$$|e^{ikx} M_{ij}|^2 = |M_{ij}|^2, \quad (\text{C.32})$$

the ideal travelling-wave intensity does not depend on the laser centre. Consequently, the current code does not spatially average the travelling wave.

C.8 Step 8: Standing-Wave Strain

The SAW wavenumber is

$$k = \frac{2\pi}{\lambda_{\text{SAW}}}. \quad (\text{C.33})$$

For

$$\lambda_{\text{SAW}} = 6 \mu\text{m}, \quad (\text{C.34})$$

the wavenumber is

$$k = \frac{2\pi}{6} \approx 1.0472 \text{ rad}/\mu\text{m}. \quad (\text{C.35})$$

The standing wave is modelled using

$$u_{xx}^{\text{SW}}(x) = 2C_{\text{SW}}u_{xx} \cos(kx), \quad (\text{C.36})$$

$$u_{zz}^{\text{SW}}(x) = 2C_{\text{SW}}u_{zz} \cos(kx), \quad (\text{C.37})$$

$$u_{xz}^{\text{SW}}(x) = 2C_{\text{SW}}u_{xz} \sin(kx). \quad (\text{C.38})$$

The normal components vary as $\cos(kx)$, whilst the shear component varies as $\sin(kx)$. This reproduces the approximate quarter-wavelength displacement observed in the lateral strain profiles.

The factor of two comes from combining the two equal counterpropagating travelling waves. It is valid only when the input strain values are single-travelling-wave amplitudes.

Here is a dimensionless normalization factor accounting for the definition of the input strain amplitudes. $C_{\text{SW}} = 1$ when they Here, C_{SW} is a dimensionless normalization factor that ensures consistency with the definition of the input strain amplitudes; $C_{\text{SW}} = 1$ when they represent one travelling wave, because two equal counterpropagating waves produce twice the single-wave amplitude at a standing-wave antinode.

The implementation is

```
def strain_operator_standing(
    a_hz,
    strain,
    spatial_factor=(
        STANDING_WAVE_SPATIAL_FACTOR
    ),
    kx_rad=STANDING_WAVE_KX_RAD,
):
    u_xx, u_zz, u_xz = strain

    standing_strain = (
        2.0 * spatial_factor
        * u_xx * np.cos(kx_rad),

        2.0 * spatial_factor
        * u_zz * np.cos(kx_rad),
```

```

        2.0 * spatial_factor
        * u_xz * np.sin(kx_rad),
    )

    return strain_operator_local(
        a_hz,
        standing_strain,
    )

```

The coordinate is measured relative to a chosen normal-strain antinode:

$$kx = \frac{2\pi}{\lambda_{\text{SAW}}}(x - x_{\text{antinode}}). \quad (\text{C.39})$$

Here, x denotes the lateral coordinate along the SAW propagation direction, measured relative to the chosen normal-strain antinode, whilst `laser_center_um` specifies the position x_0 of the centre of the Gaussian laser spot along this coordinate, in micrometres.

$$x_0 = x - x_{\text{antinode}}. \quad (\text{C.40})$$

For a 6 μm wavelength:

x_0	Normal strain	Shear strain
0 μm	Antinode	Node
0.75 μm	Mixed	Mixed
1.5 μm	Node	Antinode
3 μm	Opposite antinode	Node
6 μm	Next equivalent antinode	Node

The pattern repeats after one SAW wavelength.

C.9 Step 10: Gaussian Laser Profile

The laser illuminates a finite region rather than one exact position. Its one dimensional intensity profile along the SAW propagation direction is modelled as²

²The required analytical averages follow from the Fourier-transform pair of a Gaussian distribution [77].

$$W(x; x_0, \sigma) = \frac{1}{\sqrt{2\pi}\sigma} \exp \left[-\frac{(x - x_0)^2}{2\sigma^2} \right] \quad (\text{C.41})$$

The normalization satisfies

$$\int_{-\infty}^{\infty} W(x; x_0, \sigma) dx = 1 \quad (\text{C.42})$$

Thus the final result is an average transition strength across the illuminated region.

The laser width is entered as an intensity of Full Width at Half Maximum FWHM:

$$d_{\text{FWHM}} = 2\sqrt{2 \ln 2} \sigma. \quad (\text{C.43})$$

Therefore,

$$\sigma = \frac{d_{\text{FWHM}}}{2\sqrt{2 \ln 2}}. \quad (\text{C.44})$$

The implementation is

```
sigma_um = (
    laser_fwhm_um
    / (
        2.0
        * np.sqrt(
            2.0 * np.log(2.0)
        )
    )
)
```

For example,

$$d_{\text{FWHM}} = 1 \mu\text{m} \quad (\text{C.45})$$

corresponds to

$$\sigma \approx 0.425 \mu\text{m}. \quad (\text{C.46})$$

C.10 Step 11: Spatial Integration

At a single position, the standing-wave transition strength is

$$I_{ij}(x) = \left| \langle j | \hat{O}_{\text{standing}}(x) | i \rangle \right|^2. \quad (\text{C.47})$$

A finite laser spot covers positions with different values of $\cos(kx)$ and $\sin(kx)$. The measured ensemble strength is modeled as the laser-weighted average

$$\bar{I}_{ij} = \int_{-\infty}^{\infty} W(x; x_0, \sigma) \left| \langle j | \hat{O}_{\text{standing}}(x) | i \rangle \right|^2 dx. \quad (\text{C.48})$$

This form assumes that defects at different positions emit independently, so their intensities add.

The calculation does not use

$$\left| \int W(x) M_{ij}(x) dx \right|^2, \quad (\text{C.49})$$

because that expression would describe coherent addition of transition amplitudes from different spatial positions.

C.11 Step 12: Separating Normal and Shear Operators

The standing-wave operator can be separated into cosine and sine parts:

$$\hat{O}_{\text{standing}}(x) = \hat{O}_{\text{cos}} \cos(kx) + \hat{O}_{\text{sin}} \sin(kx), \quad (\text{C.50})$$

where

$$\hat{O}_{\text{cos}} = 2aC_{\text{SW}} [u_{xx}S_x^2 + u_{zz}S_z^2], \quad (\text{C.51})$$

and

$$\hat{O}_{\text{sin}} = 2aC_{\text{SW}} u_{xz} \frac{S_x S_z + S_z S_x}{2}. \quad (\text{C.52})$$

They are constructed in code as

```
O_cos = strain_operator_local(
    a_hz,
    (
        2.0
        * STANDING_WAVE_SPATIAL_FACTOR
        * u_xx,
```

```

        2.0
        * STANDING_WAVE_SPATIAL_FACTOR
        * u_zz,

        0.0,
    ),
)

O_sin = strain_operator_local(
    a_hz,
    (
        0.0,
        0.0,

        2.0
        * STANDING_WAVE_SPATIAL_FACTOR
        * u_xz,
    ),
)

```

For a transition $i \rightarrow j$, define

$$A_{ij} = \langle j | \hat{O}_{\cos} | i \rangle, \quad (\text{C.53})$$

$$B_{ij} = \langle j | \hat{O}_{\sin} | i \rangle. \quad (\text{C.54})$$

These are implemented as

```

amplitude_cos = scalar_from_qobj(
    eigenstates[j].dag()
    * O_cos
    * eigenstates[i]
)

amplitude_sin = scalar_from_qobj(
    eigenstates[j].dag()
    * O_sin
    * eigenstates[i]
)

```

The local transition amplitude is then

$$M_{ij}(x) = A_{ij} \cos(kx) + B_{ij} \sin(kx). \quad (\text{C.55})$$

C.12 Step 13: Expanding the Local Intensity

The local transition strength is

$$I_{ij}(x) = |M_{ij}(x)|^2. \quad (\text{C.56})$$

Substituting the normal and shear amplitudes gives

$$I_{ij}(x) = |A_{ij} \cos(kx) + B_{ij} \sin(kx)|^2. \quad (\text{C.57})$$

Expanding the absolute square we get,

$$\begin{aligned} I_{ij}(x) &= |A_{ij}|^2 \cos^2(kx) + |B_{ij}|^2 \sin^2(kx) \\ &\quad + 2 \operatorname{Re} \left[A_{ij} B_{ij}^* \right] \cos(kx) \sin(kx). \end{aligned} \quad (\text{C.58})$$

The third term represents the interference between the normal-strain and shear-strain transition amplitudes.

It can be positive, negative or zero depending on the eigenstates, magnetic field angle and relative phases of the two matrix elements.

C.13 Step 14: Analytical Gaussian Integration

Direct numerical integration over hundreds of x -positions is unnecessary because the required Gaussian averages have exact analytical solutions.

The three required averages are

$$\langle \cos^2(kx) \rangle = \int W(x) \cos^2(kx) dx, \quad (\text{C.59})$$

$$\langle \sin^2(kx) \rangle = \int W(x) \sin^2(kx) dx, \quad (\text{C.60})$$

and

$$\langle \cos(kx) \sin(kx) \rangle = \int W(x) \cos(kx) \sin(kx) dx. \quad (\text{C.61})$$

Using the characteristic function of a Gaussian distribution [77],

$$X \sim \mathcal{N}(x_0, \sigma^2) \implies \mathbb{E}[e^{iqX}] = e^{iqx_0 - q^2\sigma^2/2}, \quad (\text{C.62})$$

and taking its real part gives

$$\mathbb{E}[\cos(qX)] = e^{-q^2\sigma^2/2} \cos(qx_0). \quad (\text{C.63})$$

Setting $q = 2k$ and using $\cos^2 \theta = [1 + \cos(2\theta)]/2$, we obtain

$$\langle \cos^2(kx) \rangle = \frac{1}{2} \left[1 + e^{-2k^2\sigma^2} \cos(2kx_0) \right]. \quad (\text{C.64})$$

For a Gaussian centred at x_0 , they are

$$\langle \cos^2(kx) \rangle = \frac{1}{2} \left[1 + e^{-2k^2\sigma^2} \cos(2kx_0) \right], \quad (\text{C.65})$$

$$\langle \sin^2(kx) \rangle = \frac{1}{2} \left[1 - e^{-2k^2\sigma^2} \cos(2kx_0) \right], \quad (\text{C.66})$$

and

$$\langle \cos(kx) \sin(kx) \rangle = \frac{1}{2} e^{-2k^2\sigma^2} \sin(2kx_0). \quad (\text{C.67})$$

The quantity

$$C_{\text{spatial}} = e^{-2k^2\sigma^2} \quad (\text{C.68})$$

is the remaining spatial contrast after averaging over the laser.

It is implemented as

```
k_rad_per_um = (
    2.0 * np.pi / wavelength_um
)

kx0_rad = (
    k_rad_per_um * laser_center_um
)
```

```

spatial_contrast = np.exp(
    -2.0
    * (k_rad_per_um * sigma_um) ** 2
)

average_cos_squared = 0.5 * (
    1.0
    + spatial_contrast
    * np.cos(2.0 * kx0_rad)
)

average_sin_squared = 0.5 * (
    1.0
    - spatial_contrast
    * np.cos(2.0 * kx0_rad)
)

average_cos_sin = 0.5 * (
    spatial_contrast
    * np.sin(2.0 * kx0_rad)
)

```

C.14 Step 15: Final Averaged Standing-Wave Strength

The final laser-averaged transition strength is

$$\begin{aligned} \bar{I}_{ij} = & |A_{ij}|^2 \langle \cos^2(kx) \rangle + |B_{ij}|^2 \langle \sin^2(kx) \rangle \\ & + 2 \operatorname{Re} \left[A_{ij} B_{ij}^* \right] \langle \cos(kx) \sin(kx) \rangle \end{aligned} \quad (\text{C.69})$$

The exact code expression is

```

averaged_strength = (
    abs(amplitude_cos) ** 2
    * average_cos_squared

    + abs(amplitude_sin) ** 2
    * average_sin_squared
)

```

```

+ 2.0
* np.real(
    amplitude_cos
    * np.conjugate(amplitude_sin)
)
* average_cos_sin
)

```

A numerical round off correction is applied using

```

averaged_strength = max(
    float(np.real(averaged_strength)),
    0.0,
)

```

This removes small negative values produced by floating points. A physical intensity cannot be negative.

C.15 Step 16: Narrow and Wide Laser Limits

For a very narrow laser spot,

$$\sigma \rightarrow 0, \quad (\text{C.70})$$

so

$$e^{-2k^2\sigma^2} \rightarrow 1. \quad (\text{C.71})$$

The spatially averaged result approaches the local transition strength at the laser center:

$$\bar{I}_{ij} \rightarrow |A_{ij} \cos(kx_0) + B_{ij} \sin(kx_0)|^2. \quad (\text{C.72})$$

For a wide laser spot covering several SAW periods,

$$k\sigma \gg 1, \quad (\text{C.73})$$

and therefore

$$e^{-2k^2\sigma^2} \rightarrow 0. \quad (\text{C.74})$$

The spatial averages become

$$\langle \cos^2(kx) \rangle \rightarrow \frac{1}{2}, \quad (\text{C.75})$$

$$\langle \sin^2(kx) \rangle \rightarrow \frac{1}{2}, \quad (\text{C.76})$$

$$\langle \cos(kx) \sin(kx) \rangle \rightarrow 0. \quad (\text{C.77})$$

Thus,

$$\bar{I}_{ij} \rightarrow \frac{1}{2}|A_{ij}|^2 + \frac{1}{2}|B_{ij}|^2. \quad (\text{C.78})$$

The result then becomes almost independent of the laser centre position.

C.16 Step 17: Magnetic-Angle Sweep

The magnetic-field angle is sampled using

$$\phi_B \in [0, 2\pi]. \quad (\text{C.79})$$

The code uses

```
phi_values_rad = np.linspace(
    0.0,
    2.0 * np.pi,
    n_phi,
)
```

For `n_phi = 361`, the angular separation is approximately

$$\Delta\phi_B = 1^\circ. \quad (\text{C.80})$$

For every ϕ_B , the code:

1. constructs H_0/h ;
2. diagonalizes H_0/h ;
3. calculates each transition frequency;
4. calculates the normal-strain amplitude A_{ij} ;

5. calculates the shear-strain amplitude B_{ij} ;
6. calculates the laser-averaged standing-wave strength;
7. calculates the travelling-wave strength.

C.17 Step 18: Transition Classification

The code assigns the nominal zero-field spin ordering

$$m_s = \left(-\frac{3}{2}, -\frac{1}{2}, +\frac{1}{2}, +\frac{3}{2} \right). \quad (\text{C.81})$$

For a pair of indices i, j , it assigns

$$\Delta m_s = m_j - m_i. \quad (\text{C.82})$$

This is implemented as

```
m_values = np.array(
    [-3 / 2, -1 / 2, 1 / 2, 3 / 2]
)

delta_m = np.asarray(
    [
        m_values[j] - m_values[i]
        for i, j in pairs
    ]
)
```

The transitions are separated into nominal classes:

$$|\Delta m_s| = 1, \quad (\text{C.83})$$

$$|\Delta m_s| = 2. \quad (\text{C.84})$$

$|\Delta m_s| = 3$ transitions are omitted from the figures as the transitions of this kind are not physically observable in our experimental conditions.

At a finite transverse magnetic field, the eigenstates are generally mixtures of zero-field m_s basis states. Therefore, these Δm_s labels are nominal index-based classifications and not exact quantum numbers at every magnetic-field angle.

C.18 Step 20: Adjustable Parameters

A typical run uses

```
saved_png_files = (
    plot_all_angular_magnetic_field_strengths(
        B_T=0.05,
        temperature_K=300.0,
        wavelength_um=6.0,
        laser_center_um=0.0,
        laser_fwhm_um=1.0,
        n_phi=361,
        output_dir="angular_strength_png",
        dpi=300,
        show_plot=False,
    )
)
```

The main adjustable parameters are summarized in Table C.1.

Table C.1: Main manually adjustable parameters used in the calculation.

Code parameter	Example value	Interpretation
B_T	0.05	$B = 0.05 \text{ T} = 50 \text{ mT}$
temperature_K	300.0	$T = 300 \text{ K}$
wavelength_um	6.0	$\lambda_{\text{SAW}} = 6 \mu\text{m}$
laser_center_um	0.0	Center at the chosen normal-strain antinode
laser_center_um	1.5	Center at a shear-strain antinode for $\lambda_{\text{SAW}} = 6 \mu\text{m}$
laser_fwhm_um	1.0	Laser intensity FWHM of $1 \mu\text{m}$
n_phi	361	1° angular spacing, including both 0° and 360°
n_phi	61	Coarser 6° spacing for a quick test
dpi	300	Saved PNG resolution
show_plot	False	Save figures without displaying each plot window

Here, `wavelength_um` denotes the acoustic SAW wavelength, not the optical laser wavelength. For $\lambda_{\text{SAW}} = 6 \mu\text{m}$, the shear-strain antinode is located one quarter wavelength from the chosen normal-strain antinode:

$$\frac{\lambda_{\text{SAW}}}{4} = 1.5 \mu\text{m}. \quad (\text{C.85})$$

C.19 Step 21: Computational Improvement

The original numerical spatial calculation evaluated approximately

$$N_{\text{cases}} N_{\text{depths}} N_{\phi} N_x N_{\text{transitions}} \quad (\text{C.86})$$

matrix elements.

For four cases, four depths, 361 angles, 401 spatial points, and six transitions, this is approximately

$$4 \times 4 \times 361 \times 401 \times 6 \approx 1.39 \times 10^7 \quad (\text{C.87})$$

matrix-element calculations.

The analytical Gaussian integral eliminates the N_x loop. Only the normal and shear amplitudes must be calculated:

$$4 \times 4 \times 361 \times 2 \times 6 \approx 6.93 \times 10^4 \quad (\text{C.88})$$

matrix-element calculations.

The analytical method is therefore much faster and is exact for the assumed infinitely extended Gaussian laser profile.