Two-laser spectroscopy and coherent manipulation of color-center spin ensembles in silicon carbide

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Scientific summary

More than a century after its inception, quantum mechanics has become a well-established area of science, grounded in thorough experimental results. Extensive theoretical frameworks have been erected with the aim of harnessing the unique properties of quantum systems for making devices, for example to enable impossible-to-crack cryptography, or computers that are exponentially faster for certain tasks than what is possible now. Such a quantum computer needs many thousands of quantum systems (qubits) with their quantum states strongly intertwined, unperturbed by their environment (meaning particles, fields - anything really), yet still responding accurately to our control signals. However, quantum systems are the most fragile of things, needing great care to engineer and control, and the historically most succesful systems scale very badly from single qubit to many-qubit systems (such as ions trapped in laser cooling traps, or gaseous clouds of atoms). This scaling problem has been tackled once before for regular information technology, by designing integrated circuitry in the solid state, leading to the current age of information. The big question now is therefore: can we also build scalable *quantum* systems in the solid state? And if so, can we engineer them to be robust, operate at room temperature, and scale cheaply?

While there are many candidate solid-state qubits, the work presented in this thesis focusses on systems that naturally occur in the semiconducting material silicon carbide (SiC). SiC consists of subsequent sheets of hexagonally ordered silicon and carbon, where the sheets lie in the basal plane, and they are stacked on top of each other along the c-axis. When such a crystal of SiC is grown, even in the most thoroughly controlled environment, its atoms are not all in a perfectly regular grid: there will be defects in the lattice. The smallest of these defects, point defects on the scale of single atoms, are our systems of interest: missing atoms, substituted impurity atoms, or small complexes of such defects are examples. The electrons near such a defect locally reconfigure to minimize their energy (the ground state), upon which the combined spin (the intrinsic magnetic moment) of the electrons can, for some defects, be in several closely-

spaced quantum states. These states tend to be very stable (up to milliseconds) and coherent (the timescale within which the quantum information is preserved), and essentially form our qubit. By absorbing a photon (a particle of light) of a particular energy (the transition energy), the electrons can be made to reconfigure into higher-energy states (excited states), which in turn have a different set of spin states. This interaction with light (which for such defects in SiC tends to be in the infrared part of the electromagnetic spectrum, but also causes the sparkling of diamonds) has led to the name "color centers", and it offers an appealing way to transfer quantum information from photons to stationary qubits and back again. All these states lie deep in the forbidden gap of the semiconductor, meaning the electrons stay localized near the defect, and have very little interaction with the rest of the material - a must for a potential qubit. There are many crystallographic forms of SiC (polytypes), each with a unique array of color centers. Many of these defects all have the same orientation in the material (making for convenient operation), their operating wavelength lies on the edge of the telecom regime (convenient for integration into current-day optical networks), and the growing of SiC is an industrially mature affair. All this makes the scenario of mixing-and-matching particular color centers in SiC to applications very appealing.

Practically, when considering applications of color centers, it can be preferable to use ensembles consisting of many defects instead of single defects. Ensembles can behave as a collective single state, and with the defects spaced out along the path of the light they can benefit from collective enhancement effects due to stimulated emission. This leads to strong interaction with light, whereas single defects often require technologically demanding nano-fabrication to achieve this (e.g. optical cavities). On the other hand, due to a distribution of strain in the material, the laser energy needed to excite the electrons is not the same for all defects. Without strain, light of very particular energies is absorbed by the system (absorption lines), but this inhomogeneity causes the sharp absorption lines of differently strained defects to be smeared out into one broad line. Using a single laser resonant with the transitions, extracting information on the spin states and optical transitions is impossible, let alone use the system for quantum information applications. This effect is known as inhomogeneous broadening of the optical transitions, and is typical for color center ensembles.

This inhomogeneity is the source of the two red threads throughout this thesis. On the one hand, we develop a robust two-laser technique called spin-related emission or absorption (SRE/SRA). The key ingredient is that a homogeneous

sub-ensemble of defects has strongly enhanced interaction with light, when multiple lasers address all of its ground states at the same time. Combined with a carefully aligned static external magnetic field, this allows for detailed analysis of the spin states and optical transitions, regardless of inhomogeneous broadening. In this thesis several distinct centers in 4H-SiC are investigated with this technique. On the other hand we move beyond spectroscopy, optimizing the SRE/SRA for *coherent* interaction between the light and color centers, which would be at the core of quantum applications. By addressing the same excited state with both lasers (a Λ-system configuration) a purely quantum-mechanical destructive interference effect can take place, causing the electronic spin to remain in its ground state. This trapping of the system in its ground state is known as coherent population trapping (CPT), whereas the resulting unimpeded transmission of light through the ensemble is termed electromagnetically induced transparency (EIT). Besides providing information on the coherence times in the ensemble, EIT and CPT are essential building blocks in several proposed applications of qubits, such as quantum repeaters and memories for the quantum state of photons.

Our first investigation of the state structure of a color center in SiC is described in chapter 2. The defect under investigation is a divacancy. It consists of a carbon atom missing from the lattice, adjacent to a missing silicon atom, giving it a dumbbell shape. It is oriented along the basal plane of the material, making it lie very asymetrically in the surrounding crystal. Its ground state spin has previously been reported to consist of three spin energy states (a triplet), with small energy splittings between these states due to the low symmetry. Divacancies are naturally present in low concentration in commercially purchased wafers of 4H-SiC. To compensate for this low concentration, we use a specially designed sample from which very small amounts of light could be collected, emitted in response to direct absorption of the lasers, while filtering out the much more intense laser light (a technique called photoluminescence excitation). The resulting spectra fit very well to a simple model describing the optical excitations and decays: we confirm the ground state triplet and its splittings, and find a similar triplet structure for the excited-state spin. Identifying a Λ-system, and use it to show CPT: the first step towards all-optical quantum applications with divacancies in SiC.

In chapter 3 we explore the structure and transitions of a second type of divacancy, oriented in a more symmetrical way in the lattice, along the c-axis. Its optical transition energy is very different from the basal-plane divacancy, making

it easy to choose which type to address. The sample had been irradiated with high-energy electrons, creating a much higher defect concentration. As a result, absorption is high enough for measurements to be performed in transmission, a step towards, but not yet reaching the strong absorption (termed optical thickness) needed for actual devices. It is found that there are two distinct defects with their optical transitions overlapping, both with the ground state structure that has been attributed to the c-axis divacancy. One of these is identified as having a triplet excited state as well, whereas the excited state of the other is not conclusively identified. However, by judicious choice of the laser energies and magnetic field orientation, we can nearly exclusively have the lasers address Λ-systems. Chapter 4 proceeds with these results, showing EIT in c-axis divacancies, and exploring in-depth the influence of defect inhomogeneities and the driving of more than two transitions on EIT. The system is shown to dephase very slowly despite the optical field, showing promise for all-optical quantum control. It is also found that EIT can be robustly realized, though to be practically useful very high laser intensities are required. These are predicted to be realizable by trapping the light in very narrow optical waveguides, pointing the way for future research on these defects.

In chapter 5 we study SRE (as with the basal plane divacancies) of molybdenum interstitial color centers. These were shown to also have a triplet ground state, with splitting between the spin states almost three times larger than for divacancies, which can be useful for high-fidelity operation. No research had yet been done on the excited state (besides showing that it exists), or the coherence of the states. Contrary to existing literature, we argue our results indicate ground and excited spin states with only two levels (spin doublets), with no splittings at zero magnetic field, and anisotropic g-tensors to explain the magnetic field behaviour.