Relaxation dynamics of spin 3/2 silicon vacancies in 4H-Silicon carbide

A. J. Ramsay1 and A. Rossi2,3

1 Hitachi Cambridge Laboratory, Hitachi Europe Ltd., Cambridge CB3 0HE, United Kingdom
2 Department of Physics, SUPA, University of Strathclyde, Glasgow G4 0NG, United Kingdom
3 National Physical Laboratory, Hampton Road, Teddington TW11 0LW, United Kingdom

(Dated: March 3, 2022)

Room temperature optically detected magnetic resonance experiments on spin 3/2 Silicon vacancies in 4H-SiC are reported. The \( m_s = +1/2 \leftrightarrow -1/2 \) transition is accessed using a two microwave frequency excitation protocol. The ratio of the Rabi frequencies of the \( +3/2 \leftrightarrow +1/2 \) and \( +1/2 \leftrightarrow -1/2 \) transitions is measured to be \((0.901 \pm 0.013)\). The deviation from \( \sqrt{3}/2 \) is attributed to small difference in g-factor for different magnetic dipole transitions. Whereas a spin-1/2 system is characterized by a single spin lifetime \( T_1 \), we experimentally demonstrate that the spin 3/2 system has three distinct relaxation modes that can be preferentially excited and detected. The measured relaxation times are \((0.41 \pm 0.02)T_{\text{slow}} = T_d = (3.3 \pm 0.5)T_{\text{fast}}\). This differs from the values of \( T_p/3 = T_d = 2T_f \) expected for pure dipole (\( T_p \)), quadrupole (\( T_d \)), and octupole (\( T_f \)) relaxation modes, respectively, and implies admixing of the slow dipole and fast octupole relaxation modes.

I. INTRODUCTION

The density matrix of a qubit is often decomposed into the identity and three Pauli spin-half matrices. The resulting Bloch-vector provides an intuitive picture of the spin-half dynamics, and the populations relax with a single spin-lifetime termed \( T_1 \). A spin 3/2 system has four states, and is described by a 4x4 density-matrix. By extension, the relaxation of the four spin populations is described by three relaxation modes, characterized by three time constants. Furthermore, the 4x4 density matrix can be represented by a multipole expansion of the identity, x3 dipole (\( \mathbf{P} \)), x5 quadrupole (\( \mathbf{D} \)), and x7 octupole (\( \mathbf{F} \)) modes providing a more intuitive representation of the spin-3/2 density-matrix. This representation has advantages for understanding the spin-relaxation processes of \( S=3/2 \). For example, a dipole-like perturbation does not mix different order poles fixing the spin relaxation times such that \( T_p/3 = T_d = 2T_f \), as recently discussed theoretically in the case of a fluctuating magnetic field acting on a silicon vacancy in SiC. However, as we will demonstrate, this is not the case in practice.

An accessible spin 3/2 system for testing this prediction is the V2 silicon vacancy in 4H-SiC. Recently, a number of groups have demonstrated that defects in SiC have optically accessible spins with coherence times on a par with diamond. Unlike diamond, the manufacturing of SiC electronic devices is advanced. For example, n- and p-type doping can be routinely achieved and good quality SiO2 films can be deposited or grown on the surface, enabling CMOS processing. Due to the large breakdown voltages of SiC diodes and transistors, SiC devices are increasingly used in power electronic applications relevant to electric trains, cars and power transmission. As such, rapid improvement in materials and device quality is to be expected, and there is growing interest in SiC for quantum devices.

Here, we report room temperature optically detected magnetic resonance (ODMR) experiments on an ensemble of silicon vacancies in 4H-SiC. By using a two microwave frequency setup, the \( +1/2 \leftrightarrow -1/2 \) transition can be detected optically. Rabi oscillations of all three magnetic dipole allowed transitions are measured. The ratio of the Rabi frequencies is compared to the value of \( \sqrt{3}/2 \), expected for \( S_x \) matrix. A small difference in the in-plane g-factor for the \( +3/2 \leftrightarrow \pm 1/2 \) and \( +1/2 \leftrightarrow -1/2 \) transitions is measured. In a typical \( T_1 \) measurement a laser pulse initializes and detects the quadrupole state that decays exponentially with a time constant of \( T_d = 131 \mu s \). Here we use pulse sequences with two microwave frequencies that preferentially generate and detect the octupole and dipole states, and then measure their relaxation dynamics. The relaxation of the spin-3/2 is found to comprise of three modes with three time constants. The symmetry between exciting \( \pm 3/2 \leftrightarrow \pm 1/2 \) transitions implies that one of the relaxation modes is the quadrupole. However, the fast relaxation mode decays much faster than expected for a pure octupole relaxation mode, with \( T_{\text{fast}} < T_d/2 \). Therefore, contrary to expectations of ref, the dipole and octupole relaxation modes are admixed due to a relatively fast relaxation between \( +1/2 \leftrightarrow -1/2 \) states.

II. EXPERIMENTAL DETAILS

The silicon vacancy is a point defect due to a missing silicon atom. In 4H-SiC, there are two species of defect due to two inequivalent lattice sites with near hexagonal or cubic point symmetry. Here we nominally study an ensemble of V2 defects with a zero phonon line at 916 nm at low temperature, since it can be detected in ODMR at room temperature. The V2 is associated with the cubic k-site, and a zero-field fine structure splitting of 70 MHz.

The sample used was purchased from CREE. The substrate is n-type LPBD. There is 30 \( \mu \)m epilayer that is slightly n-type \((3 \times 10^{15} \text{ cm}^{-3})\). Due to the n-type substrate, the samples have a dark yellow color. The experiments were made using native defects of the epi-layer.
The silicon vacancy is optically polarized along the c-axis. To improve optical collection efficiency by a factor of 5-7, the chip is cleaved and mounted on its side to collect light perpendicular to the c-axis. The 785nm pump laser is chopped with an acousto-optic modulator and coupled into a home-built microscope with a dichroic mirror. The laser (14 mW) is focused on the side of the chip with a NA=0.75 air objective to a ≈ 1 μm spot-size. The photoluminescence is fiber coupled to a Si-APD module. The count-rate is set to an optimum of about 3 MHz. Typically, 10^8 counts are accumulated to achieve a standard deviation of 10^{-4}. This results in long integration times. A typical trace in fig. 1 takes about 2hrs to measure, and fig. 3(d) took about 2 weeks. The ac B-field is applied along the laser axis, perpendicular to the c-axis, using a loop antenna fashioned from a coaxial cable, and a dc B-field nominally along the c-axis is applied by positioning a permanent magnet.

III. TWO-TONE ODMR SPECTRA

![Energy-level diagram](image)

**FIG. 1.** (inset) Energy-level diagram of S=3/2 ground-state with dc B-field applied along c-axis. The magnetic dipole allowed transitions are labelled να. (main panel) (red) Single frequency ODMR spectra. ODMR is only sensitive to population difference between ±3/2 and ±1/2 states, hence the ν0 transition is not observed. (black/blue) Two frequency ODMR spectra, showing change in signal due to π-pulse inserted between two π-pulses tuned to ν1(ν2) transitions. The ν0 transition is now observed. 2f_π = 158 ± 4 MHz, f_π = 2350 ± 2 MHz. The offset is arbitrary.

The inset of fig. 1 shows the energy-level diagram of the spin-3/2 ground-state in a strong magnetic field of approximately 84 mT applied along the c-axis. To locate the transitions ν1 and ν2 a single frequency ODMR spectra is measured. This is done by applying a τ_L = 3.8 μs nonresonant laser pulse to generate a net spin in the ±3/2 state. Then a 30-ns radio frequency (rf) π-pulse is applied, if this is resonant with either the ν1 or ν2 transition the m_s = −1/2 (m_s = +1/2) state is populated resulting in a slightly increased fluorescence when a second laser pulse is applied. A lock-in measurement comparing signals with and without the rf-pulse is used. Two peaks are observed. We note that the frequency splitting 2f_π = 158 ± 4 MHz is larger than the 140 MHz expected for V_{2,S} defecst in 4H-SiC. We do not attribute this to a misalignment of the magnetic field with respect to the c-axis since this would reduce the splitting. We note that in ref[15], a slightly larger than expected splitting was also reported for single V2-related defect. Most likely, the larger than expected splitting here is due to an ensemble of silicon vacancies perturbed by nearby defects. There are a number of S=3/2 complexes associated with a negative silicon vacancy, with a nearby defect along the c-axis with various splittings[15]. In particular, the R2 complex is a close match with a splitting of 4D=157.6 MHz[15].

To detect the ν0 transition, a two frequency pulse sequence L − Mπ1,2 − T − πν − T − Mπ1,2 is used. The notation summarizes the pulse-quence with time going left to right. L indicates the laser pulse for initialization and detection. να indicates a π-pulse on the α-transition, T a time-delay. For lock-in detection, the experiment alternates between two slightly different pulse sequences at half the repetition rate. M precedes a pulse that is switched on and off at the repetition rate. Here the first π2-pulse generates a population inversion between the m_s = +1/2 and −1/2 states, that can be driven by a π0 pulse. The final π2 pulse swaps the populations of the +3/2 and +1/2 states projecting a population between ±1/2 states into the measurement basis. The two frequency spectra are displayed in fig. 1. An additional peak corresponding to ν0 is observed confirming that the ground state is spin 3/2. A dip at the ν1,2 transition is observed, since the lock-in compares the signals generated by sequences with three consecutive π1,2 and one π1,2 pulse, respectively. This is narrower than the single frequency peak, due to a spectral hole burning effect, whereby the pre-pulse selects a sub-set of defects to be probed by the πν-pulse[21]. The ν2,1 peaks are absent since the π1,2 and π2,1 pulses interact with different spin-states, and the independent unmodulated signal generated by frequency scanned πν-pulse is cancelled.

IV. RABI FREQUENCIES OF SPIN 3/2

A key property of spin-3/2 system is that the Rabi frequencies of the ν1,2 and ν0 transitions should have a ratio of √3/2 due to the ratio of the relevant elements of the S=3/2 S_z matrix. Rabi oscillation measurements are made for all three transitions, see fig. 2. Since the pre-pulse selects a sub-set of defects, the damping of the ν0 Rabi oscillation due to ensemble broadening is reduced. To measure the ratio of the Rabi frequencies, Rabi oscillations are measured as a function of rf-power at a
frequency of 2340.09 MHz by tuning the resonances with the external magnetic field. This eliminates changes in the applied power due to the frequency response of the loop-antenna. The Rabi frequencies are extracted by fit to a model that accounts for the ensemble broadening, the red-lines in fig. 2(a) give example fits. Figure 2(b) displays a plot of Rabi-frequency squared of the $\nu_2$ and $\nu_0$ transitions. This plot eliminates effects of saturation of the power applied at the loop antenna, and the intercept accounts for contributions to the effective Rabi frequency due to small detunings, or dephasing. The gradient gives $R^{-2}$, where $R$ is the ratio of the $\nu_{R2}/\nu_{R0}$ Rabi frequencies, with $R = 0.901^{+0.009}_{-0.013}$. This is larger than the value of $\sqrt{3}/2$ expected for an isotropic $S=3/2$ system.

The deviation in the ratio $R$ from $\sqrt{3}/2$ can be interpreted as a slight difference in the $g$-factors of the two transitions. We define an anisotropic $g$-factor tensor $g_{ij,k}$, such that the Zeeman Hamiltonian is $H_Z = \mu g_{ij,k} B_k S_{ij}$. The in-plane ac B-field is aligned along $x$, and $R = \frac{\sqrt{3}}{2} \frac{g_{+1/2+1/2,x}}{g_{-1/2-1/2,x}}$. Then following notation of ref.\[1\] we further define a deviation from $g_{ij,x} = g_\perp$, such that $g_{+3/2+1/2,x} = g_\perp + g_\perp$, and $g_{+1/2-1/2,x} = g_\perp - g_\perp$, and deduce $\frac{g_\perp}{g_\perp} = +0.019^{+0.005}_{-0.007}$. This is consistent with value of $\frac{g_2}{g_1} = 0.0 \pm 0.05$ reported in ref.\[1\]. For further details of the analysis, see appendix A.

V. SPIN RELAXATION MODES

The spin-relaxation dynamics of the 4-state $S=3/2$ system can be described by the matrix $\mathcal{R}$, such that $\hat{\rho}_i = -\mathcal{R}_{ij} \rho_{jj}$, where the diagonal of the density-matrix $\rho_{jj}$ represents the populations in the $m_s$ =
posed into 4 eigen-modes. The first is the identity
Therefore the spin-relaxation dynamics can be decom-
quadrupole is an eigenmode of the relaxation matrix
error. This implies a symmetry between the
cay times of $T$ with
In the special case considered in ref. with
To test this picture, a series of measurements are
To conclude, we have presented two microwave tone
data, assuming the long time limit in the signal matches
we then fit fig. 3(c) with
The data is modelled using Eq. (1). The initial state
after laser initialization is a pure quadrupole state. The
The offset is subtracted using a double lock-in method
(see appendix B), and since the signal crosses zero there
are two relaxation components with opposite sign, as expected. A single exponential fit yields $T_{1/e} = 48 \pm 7 \mu s \approx T_{1f}$, this is noticeably faster than the expected value of $T_{1f} = T_d/2 = 66 \mu s$. Figure 3(d) shows the relaxation of a mixture of the octupole and dipole modes using the pulse sequence $(L - \pi_1 \pi_0 - T - (M \pi_0) \pi_1 - L)$. After a fast initial decay, a long tail is observed, but the ratio of the fast to slow components is larger than the expected ratio of 1:1, further demonstrating admixing of the dipole and octupole relaxation modes.

VI. CONCLUSIONS
To conclude, we have presented two microwave tone optically detected magnetic resonance experiments on an ensemble of silicon vacancies in 4H-SiC, with spin 3/2. These measurements provide access to all the magnetic-dipole allowed transitions. A comparison of the Rabi frequencies for the $\pm 3/2 \leftrightarrow \pm 1/2$, and $+1/2 \leftrightarrow -1/2$ transitions allows us to measure a slightly different in-plane g-factor for these transitions. The relaxation of the
spin 3/2 system is shown experimentally to have three relaxation modes that can be preferentially generated and detected by choosing a particular microwave pulse sequence. This contrasts with a spin-1/2 system characterized by a single $T_1$-time. The spin-relaxation is approximately symmetric with respect to interchange of the ±3/2 ↔ ±1/2 transitions, indicating a pure quadrupole relaxation mode. Contrary to theory in ref[2], the decay of the short-lived octupole-like mode is faster than expected for a fluctuating in-plane B-field. This indicates mixing of the octupole and dipole relaxation modes, since a perturbation with dipole symmetry cannot mix different order poles. This suggests an additional fluctuation with quadrupole symmetry that mixes the dipole and octupole modes of odd order.[3] This may be the result of dipolar interactions with neighboring electron spin-1/2 defects, where the energy-cost of flipping a parasitic spin-1/2 matches the Zeeman splitting of the +1/2 ↔ −1/2 transition, and where the ±3/2 ↔ ±1/2 transitions are protected by an energy mismatch due to the crystal-splitting $2D \approx 70$ MHz. Or may arise due to fluctuations in the crystal-field splitting $D$. This work demonstrates that $T_1 = T_2$ measurements do not provide complete information on spin-relaxation dynamics of spin 3/2 systems.

ACKNOWLEDGMENTS

We thank the following people for their help. J. A. Haigh and R. A. Chakalov for technical assistance. Akio Shima, Kumiko Konishi and Keisuke Kobayashi of Hitachi CR for donating the material; R. Webb of EPSRC ion implantation facility at University of Surrey for C-ion implantation. G. W. Morley, H. Knowles, B. Pingault, and D. Kara for advice on ODMR measurements. A. R. acknowledges financial support from UKRI Industrial Strategy Challenge Fund through a Measurement Fellowship at the National Physical Laboratory.

Appendix A: Analysis of Rabi oscillation ratio

Because the Rabi frequency is much smaller than the splitting, $\nu_R < 25$ MHz $<< 2f_s = 158$ MHz, and the inhomogeneous broadening dominates the damping, $\Gamma^* \gg 1/T_1, 1/T_2$ we treat the system as an ensemble of detuned ideal two-level systems. The effective Rabi frequency of the transitions $i,j$[20]

$$\nu^2_{R0} = (g_{+1/2 \leftrightarrow -1/2,x} \mu_B B_{ac}(P_{rf}) )^2 + \delta_0^2, \quad (A1)$$

$$\nu^2_{R2} = (g_{+3/2 \leftrightarrow -1/2,x} \sqrt{3} \mu_B B_{ac}(P_{rf}) )^2 + \delta_2^2 \quad (A2)$$

where $\delta_i$ accounts for an error in the detuning between the rf-drive, and the transition-i, and a tiny shift due to intrinsic dephasing. $g_{i,j,k}$ is the g-factor tensor, such that the Zeeman-term in the Hamiltonian is $H_Z = \mu_B g_{i,j,k} S_{i,j,k} B_k$, where $S_{i,j,k}$ is the ij-element of $S_k$ spin-3/2 matrix. At low rf-powers ($P_{rf}$), the ac B-field $B_{ac} \propto \sqrt{P_{rf}}$. Eliminating the unknown $\mu_B B_{ac}(P_{rf})$ yields,

$$\nu^2_{R0} = \frac{2g^2_v}{3g^2_v + \nu^2_{R2} + \nu^2_{R0}} + \text{constant.} \quad (A3)$$

The red-lines in fig. 2(a) show example fits used to extract the Rabi frequencies. The Rabi oscillation signal $S(T)$ an ideal two-level system with detuning, as given by Eq. (3.16) of ref[29] is averaged over a Gaussian distribution of detunings, $\Delta$.

$$S(T) \propto \int d\Delta \frac{\Omega^2_R}{\Delta R^2} \sin^2 \left( \frac{\Lambda_R T}{2} \right) e^{-\Delta^2/\Delta^2} \quad (A4)$$

where $\Omega_R = 2\pi \nu_R$ is the Rabi frequency and $\Delta_R^2 = \Omega_R^2 + \Delta^2$ is the effective Rabi frequency. The model has three fitting parameters: the amplitude, the inhomogeneous broadening $\Delta_0$, and the Rabi frequency $\nu_R$. The gradient of fig. 2(b), gives the ratio of the $R = \nu_{R2}/\nu_{R0} = 0.901 \pm 0.007 > \sqrt{3}/2$.

To evaluate systematic errors, the ratio $R$ was calculated as a function of B-field angle, inhomogeneous broadening and E-parameter. Inhomogeneous broadening effectively dresses the Rabi frequency increasing the ratio $R$ by $\Delta R_{inhomo} \ll +0.004$. The effects of misaligned B-field, and strain are computed by considering the zero B-field Hamiltonian $H_0 = D(S_2^2 - 5/4) + E(S_1^2 - S_2^2)$, where $D = 35$ MHz, and $E$ is expected to be small[1] and an isotropic Zeeman-Hamiltonian, where $g_{ijk} = g$. The ratio $R$ increases with out-of-plane B-field, with a maximum value of $R=0.90$ at $90^\circ$. A large misalignment angle of $10^\circ$ is found to increase $R$ by $\Delta R = +0.0009$. We find that: $\Delta R_E = -3.5 \times 10^{-4}$ MHz$^{-1} E$. An upper limit of $|E| < 18$ MHz is given by the splitting between the $\nu_1$ and $\nu_2$ transitions, with $2f_s = 4\sqrt{D^2 + E^2}$[3], yielding $|\Delta R_E| < 0.006$. Combining these errors yields $R = 0.901^{+0.009}_{-0.013}$.

Appendix B: Double-lock-in method

The data collected in sec. V uses a double lock-in to achieve a stable zero offset. We use a gated APD module with 2.5 dark cps (Laser Components Count-10). The 15-ns TTL output is switched between two channels of an open-source photon counter[20] using a microwave-switch (Mini-circuits ZWSAWA-2-50DRA+). The switch slightly attenuates the TTL pulse, and it is necessary to terminate the FPGA inputs with 100$\Omega$, rather than the usual 50$\Omega$, to get reliable counting. An external 100-MHz clock is used (AEI97000CS), since the internal 48MHz clock is too slow.

In general, the rf-pulse sequence alternates between sequences S1 and S2 which are directed to channels 1 and 2. At a slower frequency, typically $\sim 0.1$Hz the order of the sequences is swapped so that S2 and S1 are directed to channels 1 and 2. By calculating $S = \frac{S_{11} - S_{22}}{S_{11} + S_{22}} - \frac{S_{21} - S_{12}}{S_{21} + S_{12}}$,
a small imbalance in the detection channels $\sim 10^{-4}$ is cancelled.

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