Supplemental Material for Telecom-band quantum memory with chlorine defects in silicon carbide

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SUPPLEMENTARY FIGURES

Supplementary Figure S1: Room-temperature time-resolved spectroscopy

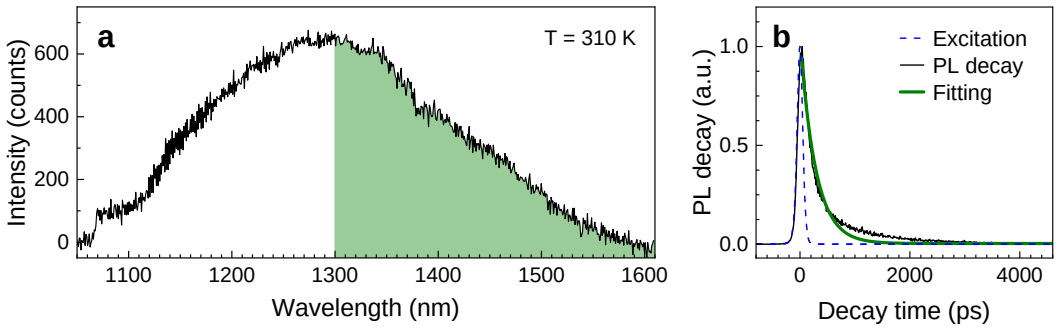

Fig. S1: (a) Room-temperature ($T = 310\,\mathrm{K}$) PL spectrum of a ^{35}Cl-implanted 4H-SiC wafer (sample #1) under optical excitation at $\lambda_{ex} = 976\,\mathrm{nm}$. The shaded area indicates the spectral range used for the PL decay measurements, selected with a longpass filter 1300 nm. (b) PL decay measurement under pulsed excitation at $\lambda_{ex} = 1060\,\mathrm{nm}$ and a pulse duration of 10 ps (the dashed line). The fitting curve represents a convolution of the laser pulse profile and a mono-exponential decay with a time constant of 265 ps.

Supplementary Figure S2: RF-assisted spectroscopy

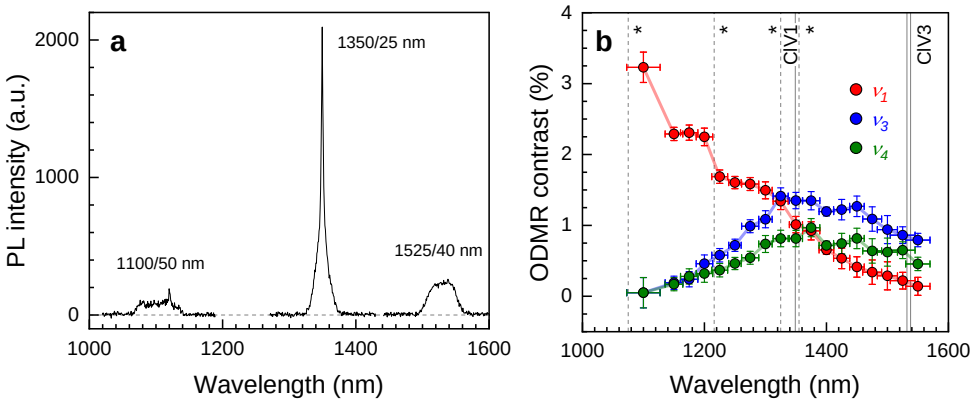

Fig. S2: (a) PL spectra measured with different bandpass filters from sample #1 under optical excitation at $\lambda_{ex} = 976\,\text{nm}$ and at temperature $T = 10\,\text{K}$. (b) ODMR contrast for the ν_1, ν_2 and ν_3 resonances detected with different bandpass filters. The vertical solid lines corresponds to the ClV1 and ClV3 ZPL. The vertical dashed lines correspond to ZPLs labeled with asterisks (*) in the main text. The vertical error bars represent standard deviation. The horizontal error bars represent the spectral detection windows.

Supplementary Figure S3: RF-induced PL dynamics

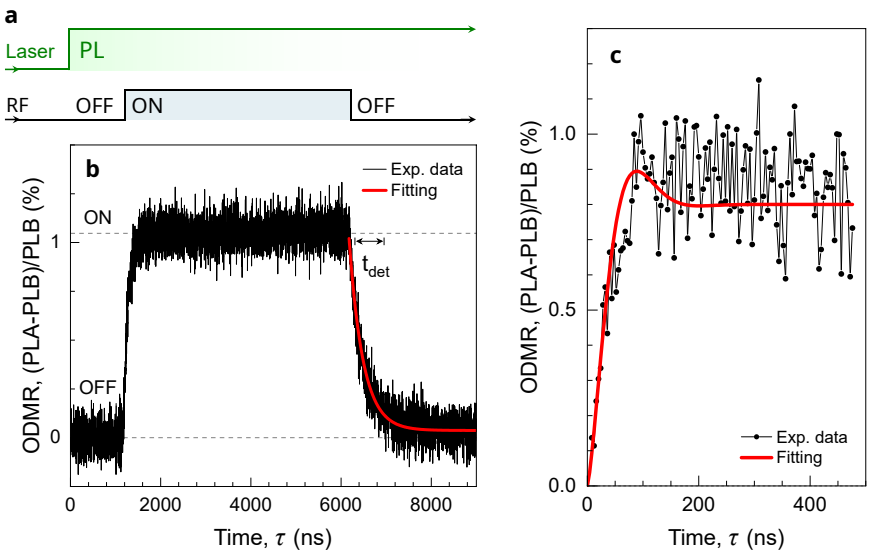

Fig. S3: (a) Pulse sequence used to measure the relative change in the PL intensity (ΔPL/PL) upon switching the RF field on and off. (b) RF-induced transient change $C = \Delta\text{PL}/\text{PL}$ in sample #1 at $T = 10\,\text{K}$. The thick red line represents a mono-exponential fit of the PL decrease after the RF field is switched off. The red thick line represents a mono-exponential fit to the PL decay after the RF field is switched off. This fit defines the detection window t_{det} used to measure PL_A and PL_B, as described in the main text. (c) Rabi measurement performed using the protocol described in the main text. The red thick line shows a fit to the function $C(\tau) = C_\infty\,(1 - \cos\omega_R\tau)\,e^{-(\frac{\tau}{T_R})^k}$ with $C = 0.8 \pm 0.1$, $\omega_R/2\pi = 4.3 \pm 0.4\,\text{MHz}$, $T_R = 50 \pm 10\,\text{ns}$ and $k = 1.2 \pm 0.3$.

Supplementary Figure S4: Ramsey fringes

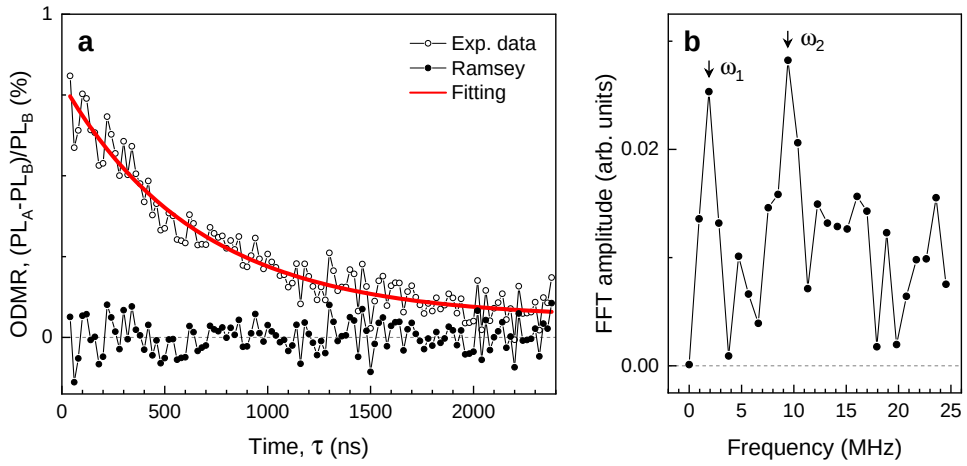

Fig. S4: (a) Open symbols represent experimental data obtained from sample #1 at $T = 10\,\mathrm{K}$ using the protocol for measuring T_2^*, as described in the main text. The red thick line shows a fit with a fixed exponential decay time determined from the T_1 measurement. Closed symbols represent the experimental data after subtraction of the exponential decay associated with quenching of the ODMR contrast. (b) Fast Fourier transform (FFT) of the experimental data after subtraction of the exponential decay. The vertical arrows indicate the frequencies of the Ramsey oscillations ω_1 and ω_2 used to fit the data in the main text.

Supplementary Figure S5: Experimental geometry

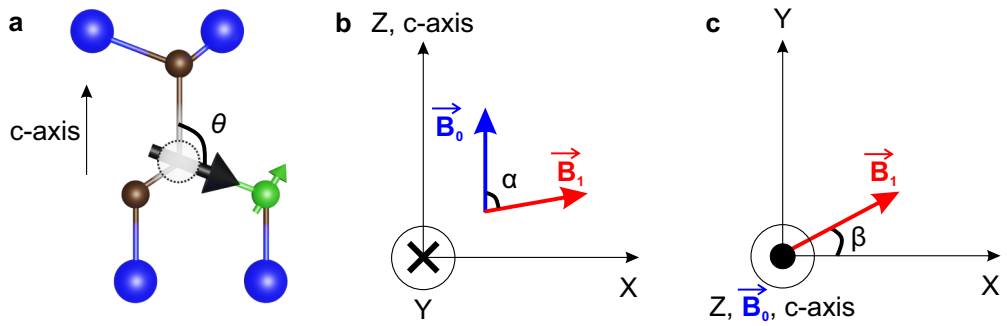

Fig. S5: (a) The 4H-SiC crystal c-axis is oriented along the Z axis of the laboratory coordinate system. Angle θ is defined as the tetrahedral angle. (b) The external magnetic field $\vec{B}_0 \parallel Z$ is oriented along the c-axis. The driving RF filed $\vec{B}_1$ is oriented at angle α with respect to the Z axis in the ZX plane. (c) The RF driving field $\vec{B}_1$ is oriented at angle β with respect to the X axis in the XY plane.

SUPPLEMENTARY TEXT

Supplementary Text S1: Simulation of the ODMR spectra

This code provides a quantitative simulation of ODMR spectrum of a spin defect in a crystalline environment, incorporating zero-field splitting, hyperfine interaction, external magnetic field and orientation effects. The code was run on MATLAB 2022b with package EasySpin 6.0.

0. Reset the work space:

```
clear;  %Removes all previously defined variables
clf;  %Clears the current figure window
```

1. Define the spin systen:

```
S = 1; %Electron spin
I = 3/2; %Nuclear spin
SiC_Cl.S = [1]; %Definition of spin system
SiC_Cl.g = [2.0028]; %Electron g factor
SiC_Cl.D = [560 60]; %Zero-field splitting parameters [D E] in MHz
SiC_Cl.Nucs = ['35Cl']; %Defiition of nuclear spin
SiC_Cl.A = -34; %Hyperfine coupling constant in MHz
SiC_Cl.lwpp = 15; %Peak-to-peak linewidth in MHz
```

2. Define the initial spin state due to optical pumping, i.e., only the $m_s = \pm 1$ spin states are assumed to be equally populated, while the population of the $m_s = 0$ spin states is zero:

```
Sz = sop([S I],'z1'); %Electronic spin projection on z
rho = abs(Sz)/8; %Density matrix rho 12 by 12
SiC_Cl.initState = {rho,'uncoupled'}; %Set initial spin state
```

3. Experiment parameters

3.1. Define crystal symmetry

```
Exp.CrystalSymmetry = 'Cs'; %Set the point group
Exp.Frame = 'crystal'; %Sample in the crystal coordinate system
```

3.2. Define static magnetic field

```
Exp.Field = 6.7 %Magnetic field in mT along z
```

3.3. Define RF driving field, with the propagation direction and polarization angle alpha (α) of the RF field explained in Fig. S5:

```
mwRange = [0 1]  %RF range in GHz
Exp.mwRange = mwRange;
Exp.Harmonic = 0; %Only fundamental harmonic
alpha = 80*pi/180; %Polarization angle
Exp.mwMode = {'y', alpha}; %RF propagation  and polarization
```

3.4. Define orientation of the lab frame, with the Euler angles beta (β) and theta (θ) for the lab frame to the sample frame transformation explained in Fig. S5:

```
beta = deg2rad(28); %Orientation of RF wire
theta = deg2rad(acosd(-(1/3)));  %Tetrahedral angle
Exp.SampleFrame = [beta theta 0]; %Set Euler angles
```

4. Define output arguments and simulation options:

```
Opt.Threshold = 1e-1; %Remove weak transitions
Opt.Verbosity = 2; %Diagnostic output
```

5. Perform simulation and plot spectra:

```
[nu, spec, info] = pepper(SiC_Cl,Exp,Opt); %Call main function
plot(nu, abs(spec), 'r'); %Plot spectrum
```

SUPPLEMENTARY TABLES

Supplementary Table S1: Spin state decomposition

TABLE S1: The decomposition is performed in the basis $|m_S, m_I\rangle$ for $B = 0\,\mathrm{mT}$. The energies are given in MHz relative to the minimum energy.

Level	Energy	$\|-1,-3/2\rangle$	$\|-1,-1/2\rangle$	$\|-1,1/2\rangle$	$\|-1,3/2\rangle$	$\|0,-3/2\rangle$	$\|0,-1/2\rangle$	$\|0,1/2\rangle$	$\|0,3/2\rangle$	$\|1,-3/2\rangle$	$\|1,-1/2\rangle$	$\|1,1/2\rangle$	$\|1,3/2\rangle$
1	0	0	0	0	0	0.01	0	0.98	0	0	0.01	0	0
2	0	0	0	0.01	0	0	0.98	0	0.01	0	0	0	0
3	4	0	0	0	0	0	0.01	0	0.98	0	0	0.01	0
4	4	0	0.01	0	0	0.98	0	0.01	0	0	0	0	0
5	489	0	0	0	0.17	0	0	0	0	0	0	0	0.83
6	489	0.83	0	0	0	0	0	0	0	0.17	0	0	0
7	508	0	0.63	0	0	0	0	0	0	0	0.36	0	0
8	508	0	0	0.36	0	0	0	0	0	0	0	0.63	0
9	632	0	0.35	0	0.02	0	0	0	0	0	0.62	0	0
10	632	0	0	0.62	0	0	0	0	0	0.02	0	0.35	0
11	648	0	0.01	0	0.81	0	0	0	0	0	0.01	0	0.17
12	648	0.17	0	0.01	0	0	0	0	0	0.81	0	0.01	0