

Oxygen-vacancy origin of robust surface-proximity spin centers in silicon carbide

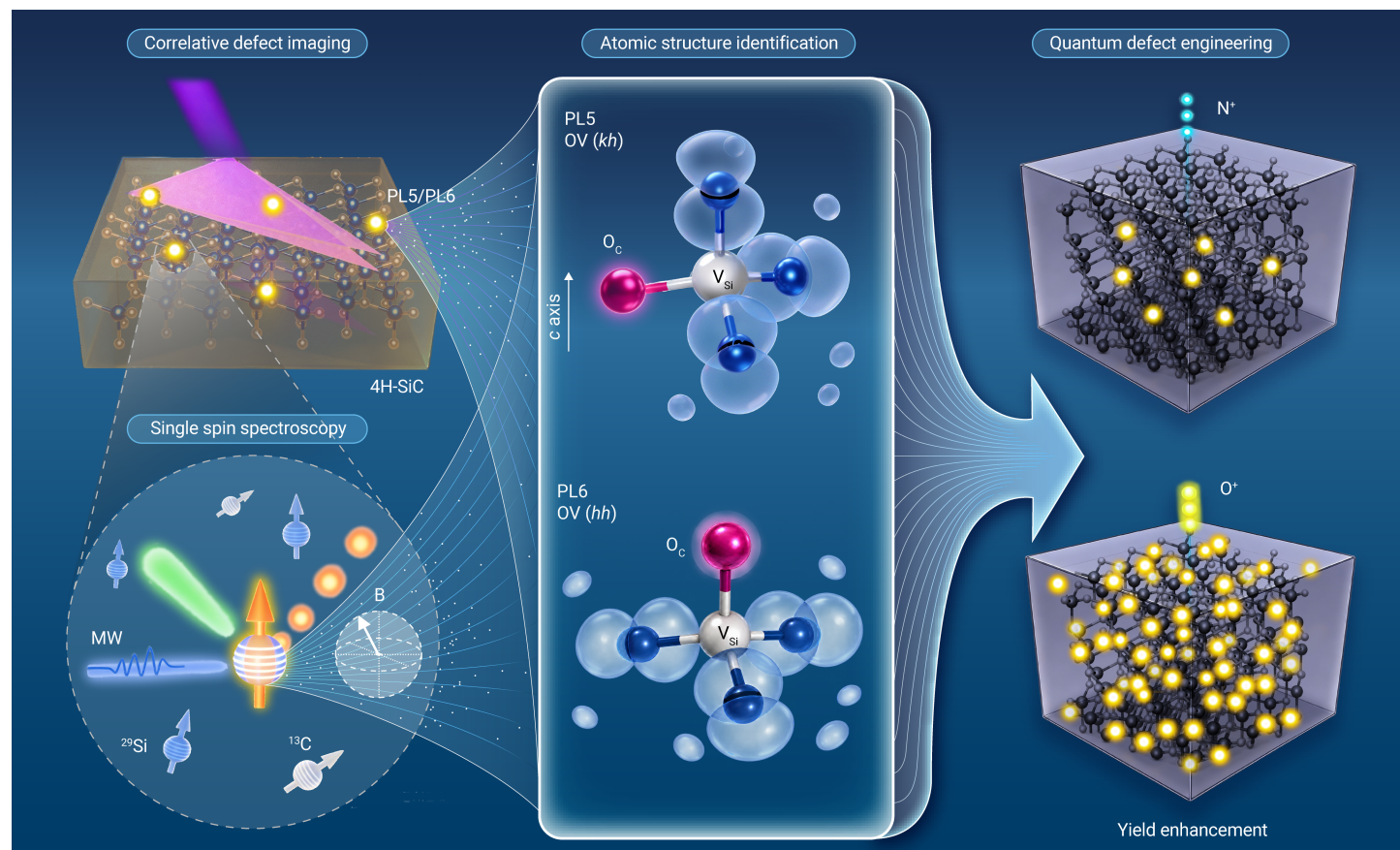
Yu Chen,^{1,2,7} Qi Zhang,^{1,3,4,7,*} Mingzhe Liu,^{1,7} Junda Wu,^{1,3,7} Jinpeng Liu,^{1,3} Xin Zhao,¹ Jingyang Zhou,¹ Pei Yu,¹ Shaochun Lin,¹ Yuanhong Teng,¹ Wancheng Yu,⁵ Ya Wang,^{1,2,6} Chang-Kui Duan,^{1,2,6} and Fazhan Shi^{1,2,3,6,*}

*Correspondence: zhq2011@ustc.edu.cn (Q.Z.); fzshi@ustc.edu.cn (F.S.)

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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- PL5 and PL6 in 4H-SiC are key quantum sensors, but their unknown structures block efficient fabrication.
- We identify both as oxygen-vacancy complexes and boost their formation yields via oxygen implantation.
- This could enable scalable SiC quantum technologies, from sensor arrays to integrated photonic devices.

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¹Laboratory of Spin Magnetic Resonance, School of Physical Sciences, and Anhui Province Key Laboratory of Scientific Instrument Development and Application, University of Science and Technology of China, Hefei 230026, China

²Hefei National Laboratory, University of Science and Technology of China, Hefei 230088, China

³School of Biomedical Engineering and Suzhou Institute for Advanced Research, University of Science and Technology of China, Suzhou 215123, China

⁴Institute of Quantum Sensing, Institute of Fundamental and Transdisciplinary Research, Zhejiang Key Laboratory of R&D and Application of Cutting-edge Scientific Instruments, State Key Laboratory of Ocean Sensing, School of Physics, Zhejiang University, Hangzhou 310027, China

⁵State Key Laboratory of Crystal Materials, Institute of Novel Semiconductors, Shandong University, Jinan 250100, China

⁶Hefei National Research Center for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei 230026, China

⁷These authors contributed equally

*Correspondence: zhq2011@ustc.edu.cn (Q.Z.); fzshi@ustc.edu.cn (F.S.)

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Spin defects promise atomic-scale resolution and single-electron sensitivity, but typically deteriorate near surfaces. The PL5 and PL6 centers in SiC have emerged as a notable exception, exhibiting high optical contrast, long spin coherence times, and stable charge states within a few nanometers of the surface at room temperature. However, deterministic creation and scalable fabrication of these centers remain limited by their unresolved microscopic origins, with PL5 and PL6 frequently discussed in connection with divacancies and stacking faults. Here, we show that PL5 and PL6 do not originate from stacking faults, as regions with and without these planar defects exhibit comparable emitter densities. Instead, oxygen implantation increases the yield by more than an order of magnitude compared to nitrogen, and isotopic implantation with ¹⁷O yields a spin-5/2 hyperfine signature in the ODMR spectra, providing direct confirmation of oxygen involvement. From the characterization of the zero-field-splitting tensor and hyperfine interactions, together with *ab initio* calculations, we assign PL5 and PL6 to O_CV_{Si} defects in the *kh* and *hh* configurations, respectively. These results establish a basis for deterministic generation of near-surface spin qubits and their scalable integration for quantum photonics and nanoscale quantum sensing.

INTRODUCTION

Optically addressable spin defects in solids have rapidly developed into a powerful materials platform for quantum technologies, enabling nanoscale detection of nuclear spins and even single biomolecules, as well as the realization of coherent spin-photon interfaces for scalable quantum networks.^{1,2} Among candidate host materials, silicon carbide (SiC) is particularly attractive owing to its wafer-scale manufacturability and the availability of spin defects exhibiting long coherence times, including divacancies (PL1–PL4), silicon vacancies, and other spin defects.^{3,4} Within this platform, the optically active PL5 and PL6 defects have emerged as especially promising. They combine favorable room-temperature spin properties with near-infrared emission suited to biosensing and telecommunication applications.⁵ Notably, PL5 and PL6 centers retain favorable spin coherence and charge-state stability even within 2 nm of the surface (7–8 atomic bilayers), a combination rarely realized in other solid-state defect systems.^{6,7} Such shallow configurations are crucial for both quantum sensing and communication, enabling strong coupling to external target systems and facilitating integration with nanophotonic structures. Indeed, both detection of electron and nuclear spins in nearby molecules and integration into nanophotonic devices have been recently demonstrated with these surface-robust spin defects,^{6,8,9} underscoring their promise.

Despite these favorable properties, the factors governing the formation of PL5 and PL6 remain incompletely understood. Attempts to generate these centers through electron irradiation reveal a puzzling host dependence: PL5

and PL6 are detected only in specific high-purity semi-insulating (HPSI) materials.^{10–12} By contrast, conventional implantation with C, N, or He ions can reproducibly generate these centers; however, the resulting yields are substantially lower than those of other spin centers in SiC.^{13,14} These unexplained phenomena have prevented the development of efficient, position-controlled fabrication protocols for these spin centers. Efforts to solve these puzzles have long been limited by a central obstacle: the microscopic origin of these defects remains unresolved. Over the past decade, competing models have been proposed, attributing these centers either to divacancies coupled to macroscopic stacking faults or to oxygen-vacancy complexes.^{15–17} However, distinct defect configurations can produce similar calculated spectroscopic signatures within the intrinsic accuracy limits of first-principles calculations, making theoretical modeling alone insufficient for a unique structural assignment. Consequently, PL5 and PL6 are still widely regarded as “unidentified” defects or classified as “modified divacancies” in the contemporary literature,^{8,18,19} leaving a critical knowledge gap that hinders rational materials engineering aimed at improving their yields and integration.

To resolve the decade-long ambiguity surrounding the nature of PL5 and PL6, we implement an integrated strategy combining correlative defect imaging, isotopic engineering, and high-resolution single-spin spectroscopy. By combining these experimental observations with *ab initio* calculations, we provide direct evidence that rules out macroscopic stacking faults as necessary structural components and supports the assignment of these defects to oxygen-vacancy centers (O_CV_{Si}) in the *kh* and *hh* configurations. This structural assignment addresses a foundational materials puzzle, providing a rational basis for the controlled engineering of high-quality, surface-proximal spin defects for scalable quantum technologies.

MATERIALS AND METHODS

The optical and spin measurements were performed at room temperature using a home-built confocal microscope, as schematically illustrated in Figure 1A. The 4H-SiC sample was mounted on a microwave coplanar waveguide and optically addressed through an oil-immersion objective, while three-axis Helmholtz coils provided vector magnetic-field control. Under 914-nm excitation, isolated emitters were resolved in the confocal photoluminescence (PL) image (Figure 1B) and identified as PL5 or PL6 based on their characteristic optically detected magnetic resonance (ODMR) spectra (Figure 1C). Second-order photon-correlation measurements of representative PL5 and PL6 centers yielded $g^{(2)}(0) < 0.5$ in both cases (Figure 1D), confirming that the observed PL originated from single emitters. Over the investigated excitation-power range, the maximum measured PL count rates reached approximately 250 kcps for PL5 and 460 kcps for PL6 (Figure 1E), demonstrating high brightness without photonic-structure enhancement. Together, these measurements establish the basic single-emitter character-

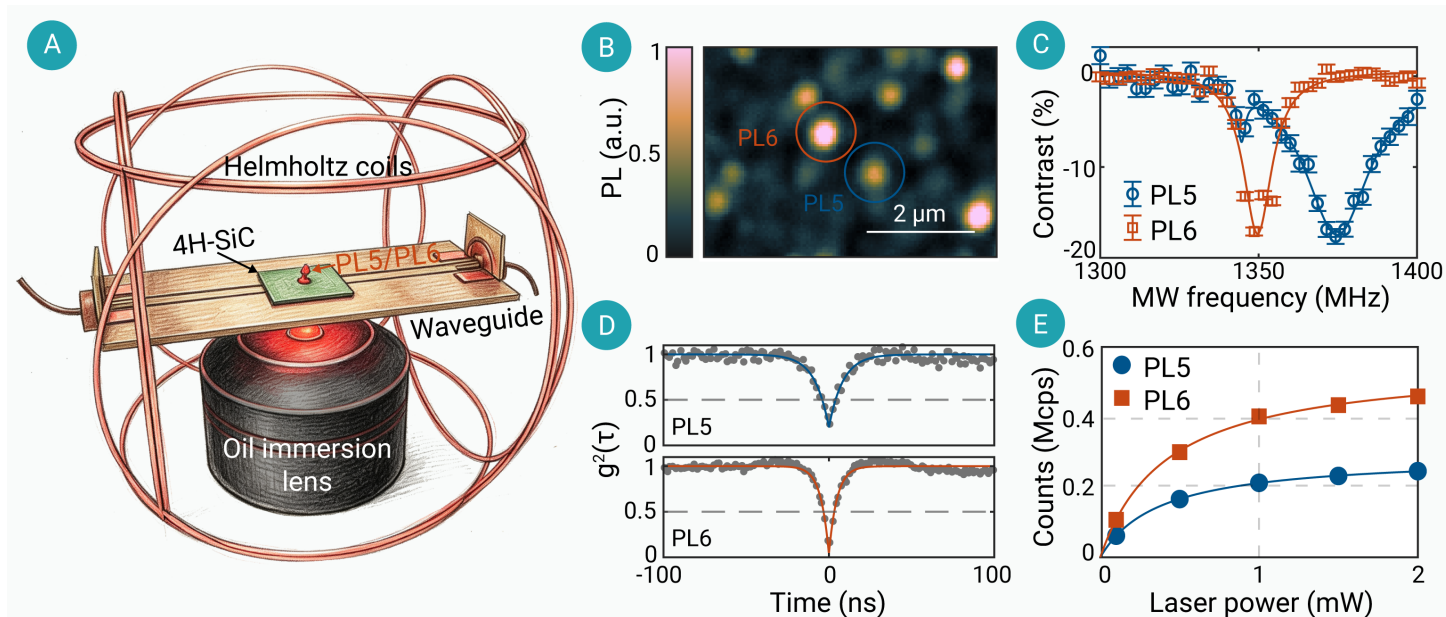


Figure 1. Experimental setup and basic optical and spin characterization of single PL5 and PL6 centers (A) Schematic of the room-temperature confocal microscope. The 4H-SiC sample is mounted on a microwave coplanar waveguide, optical excitation and collection are performed through an oil-immersion objective, and three-axis Helmholtz coils provide vector magnetic-field control. (B) Representative confocal PL image under 914-nm excitation, with individual PL5 and PL6 centers indicated. (C) Representative ODMR spectra of PL5 and PL6 centers, showing their distinct spin-resonance signatures. Symbols denote experimental data and solid curves are fits. (D) Second-order photon-correlation measurements of representative PL5 (top) and PL6 (bottom) centers. The antibunching dips with $g^{(2)}(0) < 0.5$ confirm single-photon emission. (E) Excitation-power-dependent PL count rates of single PL5 and PL6 centers. Solid curves are fits to $I(P) = I_{\text{sat}}P/(P + P_{\text{sat}})$, yielding fitted saturation count rates of $I_{\text{sat}} = 291 \pm 4$ kcps for PL5 and 558 ± 31 kcps for PL6.

istics required for the experiments described below.

RESULTS

Previous ensemble-averaged studies observed that PL5 and PL6 defects in as-grown SiC wafers are preferentially confined near the surface, leading to the prevailing hypothesis that near-surface stacking faults (SFs) are intrinsic structural components of these centers.^{15,20} Here, we test this hypothesis by probing their spatial correlation at the single-defect level, revealing a different picture: PL5 and PL6 formation does not require macroscopic stacking faults. As illustrated in Figure 2A, PL5 and PL6 centers are created in a 10^{11} cm^{-2} implantation region of Sample I via 15-keV $^{14}\text{N}^+$ implantation followed by annealing at 1000°C for 30 min in vacuum. We map the spatial locations of individual defects using ODMR, alongside the distribution of pre-existing SFs acquired via a PL-based mapping technique.²¹ Under 325-nm excitation, the presence of SFs locally reduces the bandgap, leading to a redshifted emission at 420 nm (2.95 eV) corresponding to single Shockley and intrinsic Frank SFs²¹ (see Supplemental Information, Figure S1). The spatially aligned overlay of PL5, PL6, and SF PL maps (Figure 2A) allows us to partition the area into SF-rich and SF-free regions. Representative confocal defect PL images (914-nm excitation) reveal comparable densities of PL5 and PL6 emitters in both regions (Figure 2B), providing direct evidence that their formation does not require macroscopic stacking faults.

Having ruled out macroscopic SFs as necessary structural components, we next investigate the role of oxygen incorporation, guided by recent *ab initio* calculations implicating oxygen-vacancy complexes as origins.^{16,17} Sample II was implanted with $^{16}\text{O}^+$ ions at 15 keV with a dose of 10^{11} cm^{-2} , followed by annealing at 1050°C for 30 min (Figure 2C). The confocal PL map of the oxygen-implanted regions in Figure 2D reveals a significantly higher density of emission spots. Individual PL5 and PL6 centers are again identified via ODMR (see Supplemental Information, Supp. Note2). Figure 2E quantitatively compares this $^{16}\text{O}^+$ -implanted region with a $^{14}\text{N}^+$ -implanted control region in Sample III prepared at the same implantation dose and under the same annealing conditions, and demonstrates that the formation yields of PL5 and PL6 increase by factors exceeding 11 and 19 respectively under oxygen implantation. This pronounced enhancement indicates that oxygen plays a

key role in the formation of PL5 and PL6.

To directly test oxygen incorporation, we performed isotope engineering by implanting $^{17}\text{O}^+$ ions into Sample IV (15 keV , $5 \times 10^{10} \text{ cm}^{-2}$), followed by annealing at 1050°C for 30 min. The ^{17}O nucleus possesses a nuclear spin of $I = 5/2$, which should introduce a characteristic six-fold hyperfine structure if oxygen is part of the defect core, whereas ^{16}O has $I = 0$ and should not produce such a multiplet. Consistent with this isotope dependence, the $^{16}\text{O}^+$ -implanted reference centers show no oxygen-induced hyperfine multiplet (Figure 3A), while the ODMR spectra of PL5 and PL6 in the ^{17}O -implanted sample exhibit clear six-fold hyperfine splittings (Figure 3B). The ^{17}O spectra are well fitted by six Lorentzian profiles, yielding axial hyperfine couplings A_z of $2.50(2) \text{ MHz}$ for PL5 and $3.32(2) \text{ MHz}$ for PL6. Furthermore, by increasing the aligned magnetic field, we observe a pronounced asymmetric intensity distribution among the hyperfine lines for PL6 (Figure 3C). This magnetic-field-dependent asymmetry is a hallmark of dynamic nuclear polarization (DNP), further supporting strong hyperfine coupling to the incorporated ^{17}O nucleus. Collectively, these isotope-dependent signatures provide direct spectroscopic evidence for oxygen involvement in the atomic core of both PL5 and PL6.

With oxygen incorporation established, the $\text{O}_\text{C}\text{V}_\text{Si}$ complex emerges as the leading structural candidate among the oxygen-related defects. Systematic *ab initio* screenings of various oxygen-related defects in 4H-SiC show that only the neutral $\text{O}_\text{C}\text{V}_\text{Si}$ complex simultaneously possesses the requisite $S = 1$ ground state and near-infrared zero-phonon line (ZPL) energies matching the well-established experimental signatures of PL5 and PL6.^{22,23} Consequently, PL5 and PL6 can be attributed to distinct configurations of this defect. Specifically, the $\text{O}_\text{C}\text{V}_\text{Si}$ complex in 4H-SiC can adopt four distinct configurations—*hh*, *kk*, *hk*, and *kh*—depending on whether the oxygen atom and the silicon vacancy reside in hexagonal (*h*) or quasi-cubic (*k*) lattice sites. For PL6, our direct spectroscopic identification of oxygen, together with prior ZFS and hyperfine characterizations,^{16,17} allows us to assign it to the axial *hh* configuration. The corresponding atomic structure is shown in Figure 3D. In contrast, the structural assignment of PL5 has remained controversial because key experimental constraints were missing. In particular, the orientation of the primary principal axis and the local hyperfine-coupling pattern

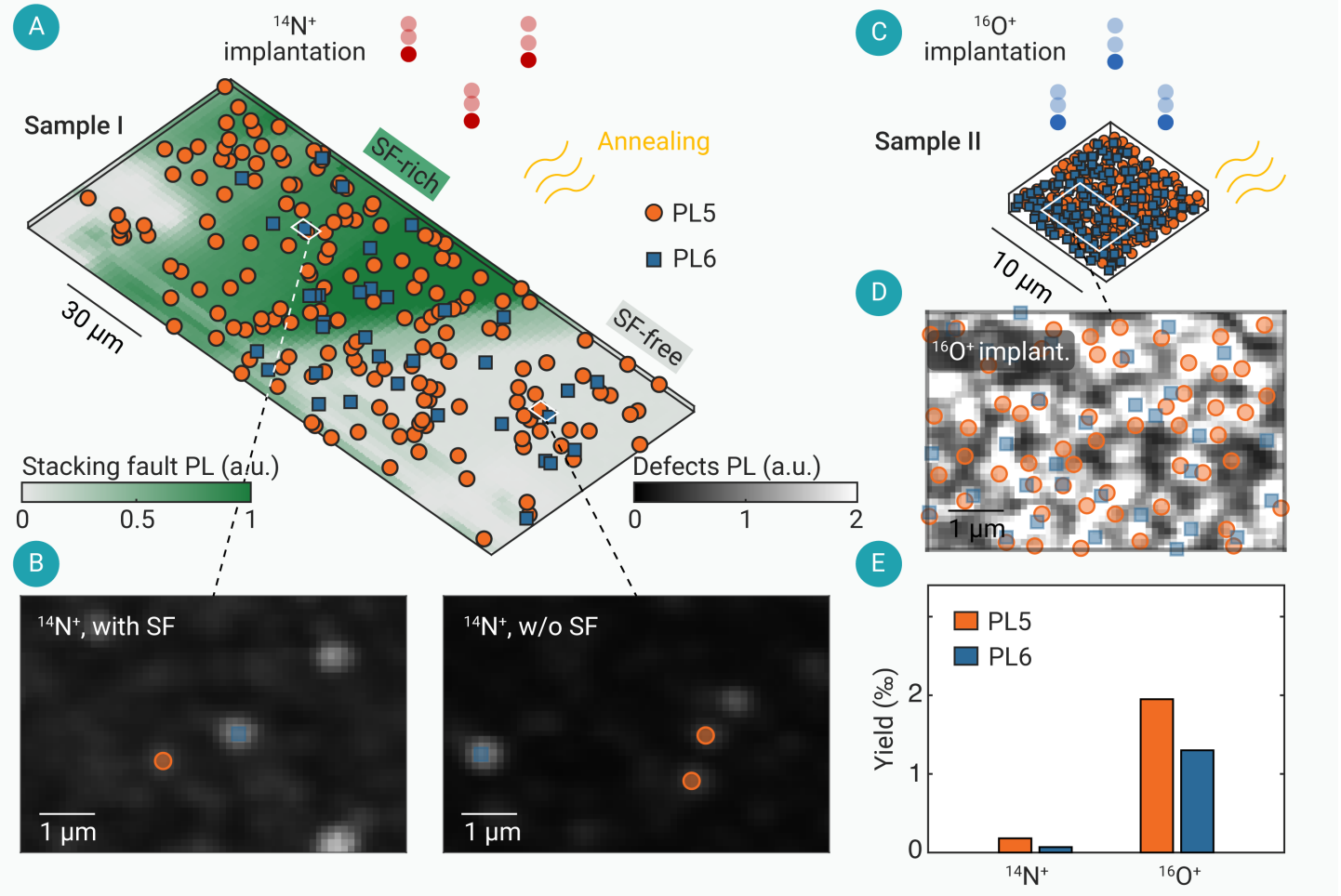


Figure 2. Decoupling PL5 and PL6 defects from stacking faults and validating oxygen-enhanced formation (A) Spatially correlated map of individual PL5 (orange circles) and PL6 (blue squares) defects against the stacking-fault (SF) background (green gradient) in Sample I, generated in a 10^{11} cm^{-2} $^{14}\text{N}^+$ implantation region followed by annealing at 1000°C for 30 min. The SF photoluminescence (PL) map is obtained by integrating emission over the 410–430 nm spectral range (325-nm excitation). Defect locations are mapped via optically detected magnetic resonance (ODMR) (see Supplemental Information, Supp. Note1). (B) Representative confocal PL images (914-nm excitation, 980–1200 nm detection) acquired in SF-rich (left) and SF-free (right) regions of Sample I, demonstrating comparable single-defect densities independent of the local SF environment. (C) Schematic of $^{16}\text{O}^+$ implantation in Sample II (15 keV, 10^{11} cm^{-2}), followed by annealing at 1050°C for 30 min. (D) Confocal PL image of the oxygen-implemented Sample II, revealing a markedly enhanced emitter density. ODMR-identified PL5 and PL6 centers are again denoted by orange circles and blue squares (see Supplemental Information, Supp. Note2). (E) Statistical comparison of the defect formation yield efficiencies between Sample II ($^{16}\text{O}^+$) and Sample III ($^{14}\text{N}^+$), both implanted at 15 keV with a dose of 10^{11} cm^{-2} and annealed at 1050°C for 30 min. The introduction of oxygen increases the generation yield of both PL5 and PL6 by more than an order of magnitude compared to nitrogen implantation.

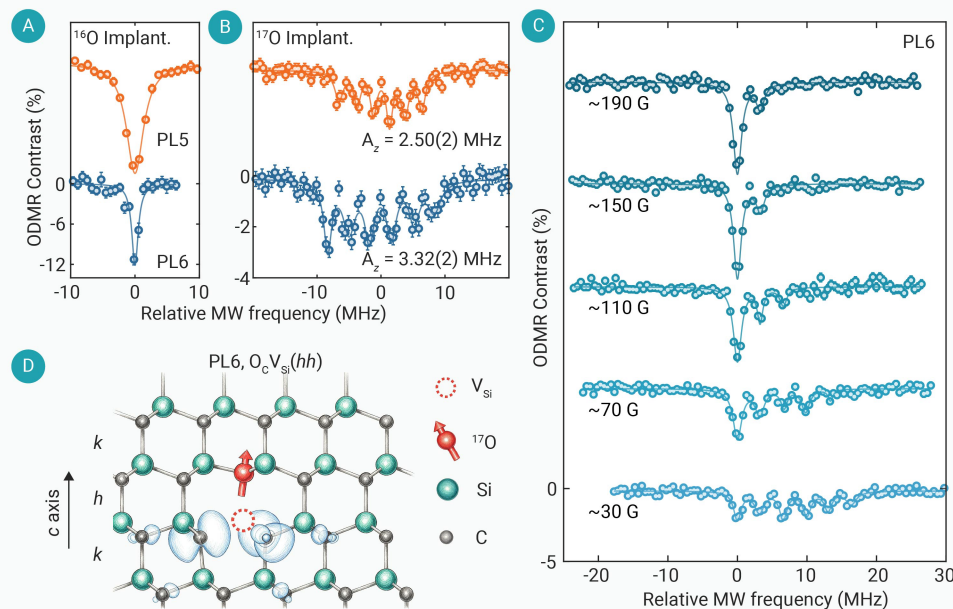


Figure 3. Direct spectroscopic evidence of oxygen incorporation via ^{17}O isotope engineering (A) Reference ODMR spectra of single PL5 (top) and PL6 (bottom) centers created by $^{16}\text{O}^+$ implantation. Because ^{16}O has nuclear spin $I=0$, no oxygen-induced hyperfine multiplet is observed. (B) ODMR spectra of single PL5 (top) and PL6 (bottom) centers measured in Sample IV, prepared by 15 keV $^{17}\text{O}^+$ implantation at a dose of $5 \times 10^{10} \text{ cm}^{-2}$ followed by annealing at 1050°C for 30 min. The external magnetic field is aligned with each defect's primary principal axis. The emergence of a characteristic six-fold splitting confirms the hyperfine interaction with the $I = 5/2$ nuclear spin of ^{17}O . (C) Magnetic field dependence of the PL6 ODMR spectrum. As the aligned magnetic field increases from $\sim 30 \text{ G}$ to $\sim 190 \text{ G}$, a pronounced asymmetric intensity distribution develops among the hyperfine transitions, indicative of dynamic nuclear polarization (DNP) of the ^{17}O nucleus. (D) Atomic model of the identified axial hh configuration corresponding to the PL6 defect. Green and gray spheres represent Si and C atoms, respectively. The substituted oxygen isotope (^{17}O) and the adjacent silicon vacancy (V_{Si}) are denoted by the red arrow and dashed circle. The calculated ground-state spin density isosurface (blue) is overlaid, and the h/k stacking sequence along the c axis is indicated.

Primary principal axis

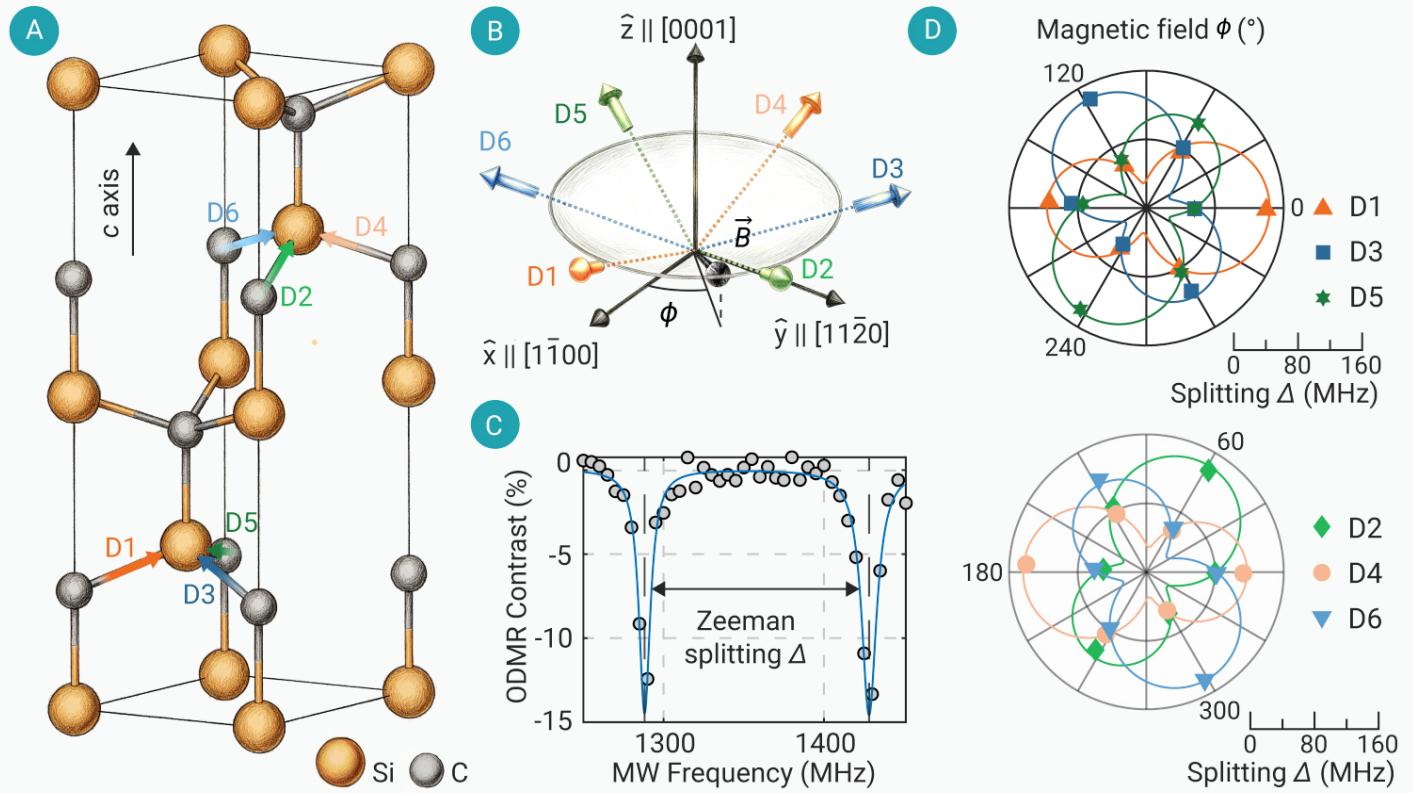


Figure 4. Determination of the six primary principal axes of PL5 (A) Schematic of the 4H-SiC lattice illustrating the six basal C–Si bond directions (D1–D6). (B) Measurement geometry for determining the primary principal axes. A static magnetic field ($B = 25$ G) is rotated along a conical surface with an azimuthal angle ϕ defined relative to the $[1\bar{1}00]$ direction. (C) Representative ODMR spectrum of a PL5 center, illustrating the Zeeman splitting. (D) Polar plots of the ODMR splittings as a function of ϕ . The observation of six distinct angular dependencies matches the D1–D6 bond directions within experimental uncertainty, supporting the assignment of PL5 to a basal configuration.

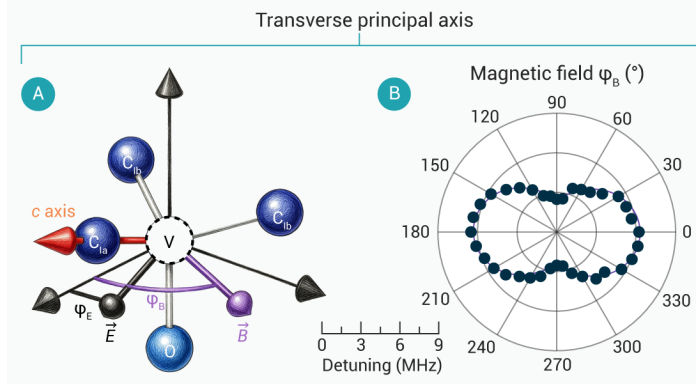


Figure 5. Transverse principal-axis determination for PL5 (A) Coordinate system for the transverse mapping. The expected mirror symmetry plane is defined as the xz -plane, encompassing the primary principal axis (z -axis), the vacancy (V), and the specific nearest-neighbor carbon atom (C_{1a}). Within the transverse xy -plane (shaded gray), the transverse ZFS anisotropy is represented by the two-component parameter $\vec{E}$ with a phase angle φ_E . A transverse magnetic field $\vec{B}$ is rotated by an azimuthal angle φ_B measured from the x -axis. (B) Polar plot of the ODMR transition detuning as a function of φ_B . Fitting the characteristic angular dependence yields the phase angle $\varphi_E = 182.2^\circ \pm 3.5^\circ$. In the mirror-plane-defined coordinate frame, this corresponds to $E_y \approx 0$, indicating that the transverse ZFS tensor is diagonal in this frame. Equivalently, one transverse principal axis is normal to the mirror plane and the other lies within it, consistent with the C_{1h} symmetry expected for a basal $O_C V_{Si}$ defect.

single primary principal axis, whereas basal configurations (hk, kh) allow six. Although previous studies reported only three primary principal axes for PL5,^{5,13,24} our precise angular ODMR measurements resolve three additional axes, confirming a total of six. As illustrated in Figures 4A–B, by rotating the azimuthal angle ϕ of a 25 G magnetic field applied along a conical surface, we observe six distinct angular dependencies (D1–D6) in the ODMR splitting (Figures 4C–D), each corresponding to a unique primary principal axis. Further absolute orientation measurements (see Supplemental Information, Supp. Note3) confirm that these six primary principal axes match the six basal C–Si bond directions in the 4H-SiC lattice within experimental uncertainty, supporting the assignment of PL5 to a basal configuration.

Having established the six primary principal axes of PL5, we next probe its transverse ZFS tensor to determine whether the measured anisotropy is consistent with the C_{1h} symmetry expected for a basal $O_C V_{Si}$ complex. We perform this measurement in the local frame of a selected center, with the z -axis aligned with its primary principal axis and the x -axis chosen within the expected mirror plane. The expected mirror plane thus coincides with the xz -plane, while the y -axis is normal to it (Figure 5A). The effective ground-state spin Hamiltonian relevant to this measurement is

$$\frac{H}{h} = D \left(S_z^2 - \frac{2}{3} \right) - E_x (S_x^2 - S_y^2) + E_y (S_x S_y + S_y S_x) + \gamma \vec{B} \cdot \vec{S}, \quad (1)$$

where D is the axial ZFS parameter, $\gamma = g\mu_B/h$, and the transverse components are parameterized as $E_x = E \cos \varphi_E$ and $E_y = E \sin \varphi_E$, with E denoting the magnitude of the transverse ZFS anisotropy. Under reflection in the xz -plane, the operator $S_x^2 - S_y^2$ is mirror-even, whereas $S_x S_y + S_y S_x$ is mirror-odd. Therefore, C_{1h} symmetry requires E_y to vanish while allowing a finite E_x . To

had not been fully resolved. As a result, different theoretical models could not be quantitatively distinguished. We address this gap through comprehensive ZFS and hyperfine measurements.

The $O_C V_{Si}$ model predicts that axial configurations (hh, kk) allow only a

Table 1. Theoretical benchmarking and structural assignment based on fine-structure and spectroscopic parameters.

Label	ZPL (eV)	D (MHz)	E_x (MHz)	$ E_y $ (MHz)	$\delta(C_{1a}) - \delta(C_{1b})$ (MHz)
(Exp.) PL5	1.189	1373	-16.5	<0.5	0.88
(Cal.) $O_C V_{Si}(kh)$	1.12	1552 (1322)	-47.0 (-47.6)	7.7	0.89
(Cal.) $O_C V_{Si}(hk)$	1.06	1440 (1215)	-96.5 (-96.9)	7.8	2.32
(Exp.) PL4	1.118	1334	$(E_x^2 + E_y^2)^{1/2} = 18.7$		<3
(Cal.) $V_C V_{Si}(kh)$	1.12	1465	-33.9	0.0	1.72
(Exp.) PL3	1.119	1222	$(E_x^2 + E_y^2)^{1/2} = 82$		9.3
(Cal.) $V_C V_{Si}(hk)$	1.08	1402	-63.9	0.0	4.57

Theoretical D values for $O_C V_{Si}$ are listed as uncorrected (spin-decontamination-corrected²⁶); divacancy D values are uncorrected and serve as computational benchmark only. Comparison of the first-principles calculated properties of divacancies (PL3, PL4) and oxygen-vacancy complexes [$O_C V_{Si}(kh)$, $O_C V_{Si}(hk)$] with experimental values. The well-characterized PL3 and PL4 centers serve to establish the baseline accuracy and calibrated error margins of our computational framework. The experimental zero-phonon line (ZPL) energies, axial ZFS parameters (D), and total transverse anisotropy magnitudes (E) for PL3, PL4, and PL5 are taken from Refs.^{24,27}. The parameters $\delta(C_{1a})$ and $\delta(C_{1b})$ represent the characteristic hyperfine splittings induced by the nearest-neighbor ^{13}C nuclei. Their splitting differences, $\delta(C_{1a}) - \delta(C_{1b})$, for PL3 and PL4 are derived from Ref.²⁸, whereas the value for PL5 is directly extracted from the statistical single-defect measurements performed in this work (Figure 6C). To test the point-group symmetry of PL5, its transverse anisotropy is resolved into specific components ($E_x = E \cos \varphi_E$, $E_y = E \sin \varphi_E$) by combining the reported magnitude of E with the phase angle φ_E determined in Figure 5B. The best overall agreement among the tested structural models supports the kh configuration for PL5.

test this symmetry-imposed condition experimentally, we follow the angular protocol established in Ref.²⁵. Specifically, we rotate a transverse magnetic field of fixed magnitude $B_{\perp} = 25$ G in the xy -plane, with φ_B denoting its azimuthal angle, and record the ODMR transition frequency $f_{\perp}$ as a function of φ_B . We then extract φ_E by fitting the resulting angular dependence to the transition-frequency expression given in Supp. Note3.2 of the Supplemental Information. As shown in Figure 5B, the fit yields $\varphi_E = 182.2^\circ \pm 3.5^\circ$, corresponding to an $E_x/E = \sin \varphi_E$ that is consistent with zero within experimental uncertainty. The measured transverse ZFS tensor is therefore consistent with the C_{1h} symmetry expected for a basal $O_C V_{Si}$ complex.

Finally, as a crucial step toward identifying the specific basal configuration (kh vs. hk), we mapped the nuclear-spin environment of PL5. As shown in Figure 6A, we first probed the strong, nearest-neighbor interactions by measuring the characteristic hyperfine splitting δ induced by adjacent ^{13}C nuclear spins (C_{1a} and C_{1b}). Figures 6B-C show that the C_{1a} -coupled centers exhibited an average splitting difference, $\delta(C_{1a}) - \delta(C_{1b})$, of 0.88 MHz. To move beyond nearest-neighbor interactions and capture the broader nuclear-spin environment, Figure 6D provides a statistical dataset of 128 individually measured PL5 centers coupled to more distant nuclear spins (^{29}Si , ^{13}C), from which we extracted the axial hyperfine couplings A_z (see Supplemental Information, Supp. Note4, for details). The resulting statistical distribution of hyperfine couplings to the surrounding nuclear-spin environment in Figure 6E sets the stage for comparison with theoretical predictions.

The assignment of the basal configuration was achieved through cross-validation between experiment and theory, combining the newly mapped hyperfine signatures (Figure 6) with the defect's established fine-structure (D , E) and zero-phonon line (ZPL) parameters (Table 1). To assess the accuracy of our first-principles calculations, we benchmarked them against the well-characterized neutral divacancies PL3 [$V_C V_{Si}(hk)$] and PL4 [$V_C V_{Si}(kh)$]. The benchmark yields error bounds of $\Delta D < 200$ MHz, $\Delta E < 30$ MHz, and $\Delta ZPL < 0.05$ eV for the uncorrected ZFS values. Applying the spin-decontamination correction of Biktagirov *et al.*²⁶ further brings the calculated D parameters of the $O_C V_{Si}$ configurations to within 50 MHz of experiment.

Within these calibrated bounds, the kh model shows better overall agreement with the experimental D , ZPL energy, and nearest-neighbor hyperfine splitting difference than the hk model (Table 1). More decisively, its transverse anisotropy E is substantially smaller than that predicted by the hk model and is therefore much closer to the experimental value for PL5. The kh model also reproduces the broad statistical profile of the extended nuclear-

spin environment (Figure 6E), whereas the hk prediction deviates substantially. Together, these spectroscopic and computational results support the assignment of PL5 to the kh basal configuration (Figure 6F), revising the previous tentative hk assignment.¹⁶

DISCUSSION

In summary, we have resolved the long-standing structural ambiguity surrounding the PL5 and PL6 centers in 4H-SiC, assigning PL5 and PL6 to the basal kh and axial hh configurations of the $O_C V_{Si}$ complex, respectively. While isotope-controlled implantation provided direct spectroscopic evidence confirming the incorporation of oxygen, the combined analysis of tensor symmetries and hyperfine interactions, along with *ab initio* calculations, supports these structural assignments.

Their assignment to the $O_C V_{Si}$ structure identifies them as point defects formed through a vacancy-impurity pairing mechanism analogous to that of the NV center in diamond. This mechanistic similarity implies that established spatial control and self-aligned patterning techniques developed for NV centers²⁹ can now be directly adapted for SiC. Consequently, this assignment provides a route toward position-controlled placement of PL5 and PL6 centers within photonic nanostructures, marking a key step toward realizing efficient spin-photon interfaces for scalable quantum networks.

Beyond nanoscale integration, our findings establish a rational framework for generating high-quality macroscopic defect ensembles. While our oxygen-implantation experiment already enhances the PL5/PL6 formation yield by more than an order of magnitude, the relevant fabrication parameters can now be systematically optimized. For instance, combining targeted oxygen incorporation with electron irradiation offers a promising top-down route for generating large-scale, three-dimensional defect ensembles. Furthermore, analogous to strategies developed for divacancies,^{30,31} a deeper understanding of the thermodynamic and charge-state conditions that maximize the $O_C V_{Si}$ yield will guide the optimization of doping protocols, implantation doses, annealing temperatures, and Fermi-level control. Such targeted materials engineering holds the potential to elevate the ensemble ODMR contrast from typical sub-percent levels toward the intrinsic single-defect limit exceeding 20%,^{13,27} a crucial leap for high-sensitivity ensemble quantum sensing.

Ultimately, the assignment of PL5 and PL6 to oxygen-vacancy complexes bridges a critical gap in solid-state defect physics. Combined with the wafer-scale manufacturability and device-processing compatibility of 4H-SiC, this knowledge paves the way toward the scalable realization of high-perfor-

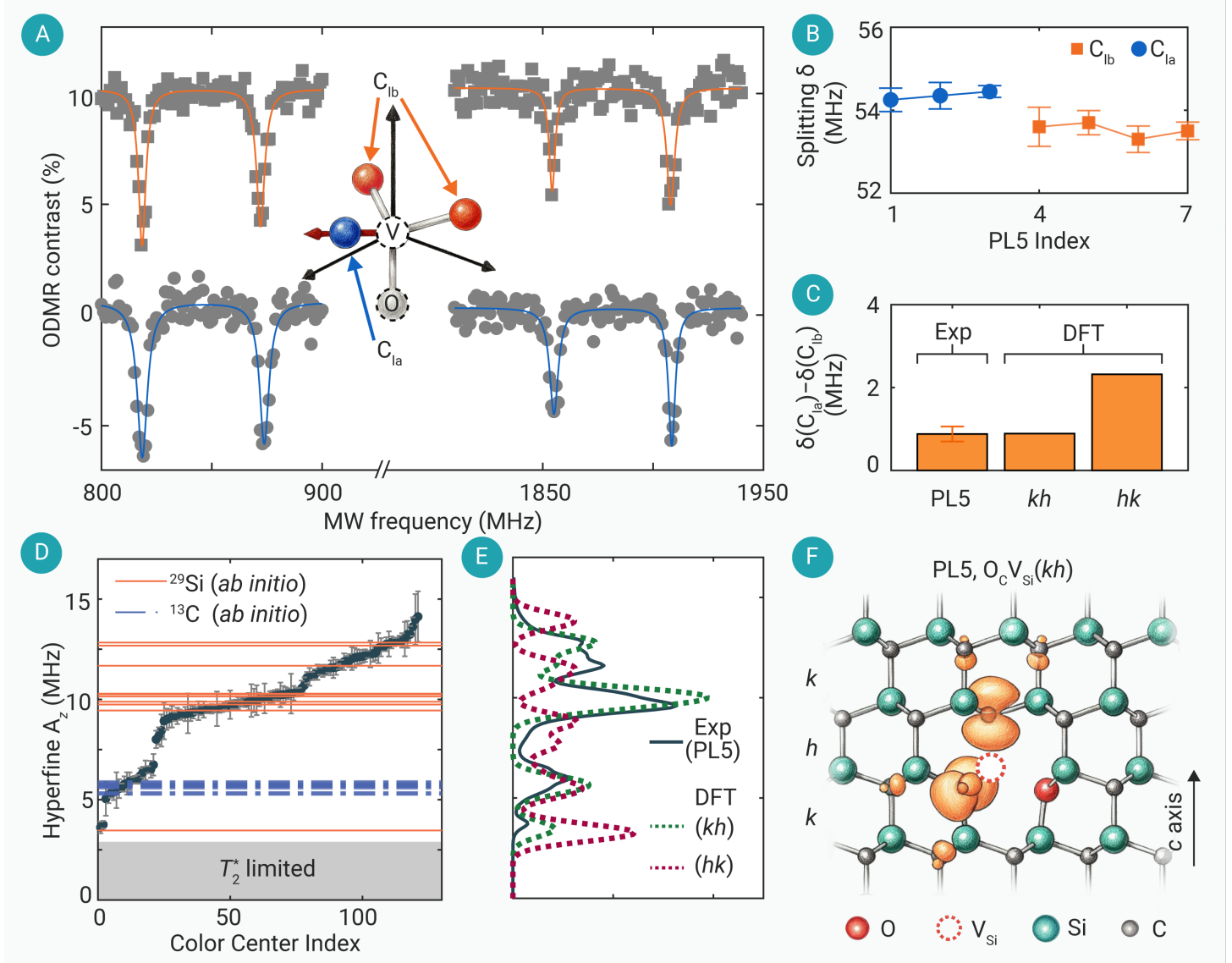


Figure 6. Statistical mapping of the local nuclear-spin environment and structural assignment of PL5 (A–C) Probing strong nearest-neighbor interactions. (A) ODMR spectra of individual PL5 centers strongly coupled to a single nearest-neighbor ^{13}C nuclear spin (C_{1a} or C_{1b}) adjacent to the silicon vacancy V_{Si} , measured at $B = 185$ G. Solid lines are multi-peak Lorentzian fits. (B) Extracted characteristic hyperfine splittings (δ) for three C_{1a} -coupled (blue circles) and four C_{1b} -coupled (red squares) PL5 centers. (C) Experimental average splitting difference, $\delta(C_{1a}) - \delta(C_{1b})$, compared with *ab initio* predictions for the *kh* and *hk* basal configurations of the $\text{O}_\text{C}V_{\text{Si}}$ defect, showing better agreement with the *kh* model. (D–E) Mapping the extended nuclear-spin environment. (D) Sorted distribution of the axial hyperfine coupling constants (A_z) extracted from a statistical dataset of 128 individually measured PL5 centers (measured at $\gamma B_2 = 39.6 \pm 1$ MHz). Couplings below 3 MHz are obscured by the intrinsic ODMR linewidth (T_2^* limited, gray shaded area). Solid orange and dashed blue lines represent the *ab initio* predicted couplings for the $\text{O}_\text{C}V_{\text{Si}}(kh)$ model. (E) Comparison between the experimental hyperfine-coupling A_z distribution (solid line) and theoretical predictions (dotted lines). The experimental profile is generated by summing Gaussian line shapes broadened by the measurement uncertainty in (D), while theoretical profiles are broadened identically. (F) Final atomic model of the identified basal *kh* configuration corresponding to the PL5 defect.

mance quantum sensors and integrated quantum communication platforms based on these versatile defects, leveraging their unique robustness at near-surface interfaces.

DECLARATION OF AI AND AI-ASSISTED TECHNOLOGIES IN THE WRITING PROCESS

During the preparation of this work the authors used ChatGPT and DeepSeek in order to improve language and readability and to assist with manuscript formatting. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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AUTHOR CONTRIBUTIONS

F.S. supervised the project. F.S. and Q.Z. proposed the idea and designed the experiments. Y.C., J.Z., J.L., and Q.Z. prepared the setup. Y.C., J.W., J.L., Y.T., and Q.Z. performed the experiments. M.L., X.Z., Y.C., and C.-K.D. carried out the DFT calculations. P.Y., Y.C., S.L., and Y.W. prepared the SiC samples. W.Y. suggested the PL mapping of stacking faults. Y.C., Q.Z., M.L., J.L., X.Z., C.-K.D., and F.S. wrote the manuscript. All authors discussed and analyzed the data, contributed to the manuscript, and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND MATERIALS AVAILABILITY

Data are available from the corresponding author upon reasonable request.