Purcell Enhancement of a Single Silicon Carbide Color Center with Coherent Spin Control

Alexander L. Crook, Christopher P. Anderson, Kevin C. Miao, Alexandre Bourassa, Hope Lee, Sam L. Bayliss, David O. Bracher, Xingyu Zhang, Hiroshi Abe, Takeshi Ohshima, Evelyn L. Hu, and David D. Awschalom*

ABSTRACT: Silicon carbide has recently been developed as a platform for optically addressable spin defects. In particular, the neutral divacancy in the 4H polytype displays an optically addressable spin-1 ground state and near-infrared optical emission. Here, we present the Purcell enhancement of a single neutral divacancy coupled to a photonic crystal cavity. We utilize a combination of nanolithographic techniques and a dopant-selective photoelectrochemical etch to produce suspended cavities with quality factors exceeding 5000. Subsequent coupling to a single divacancy leads to a Purcell factor of $\sim 50$, which manifests as increased photoluminescence into the zero-phonon line and a shortened excited-state lifetime. Additionally, we measure coherent control of the divacancy ground-state spin inside the cavity nanostructure and demonstrate extended coherence through dynamical decoupling. This spin-cavity system represents an advance toward scalable long-distance entanglement protocols using silicon carbide that require the interference of indistinguishable photons from spatially separated single qubits.

KEYWORDS: Silicon carbide, divacancy, single spin defect, Purcell enhancement, coherent spin control, photonic crystal cavity

Silicon carbide (SiC) is a technologically mature semiconductor used in commercial applications ranging from high-power electronics to light-emitting diodes. These commercial uses have led to well-developed wafer-scale fabrication processes and precise control of doping during single-crystal growth. Concurrently, SiC has generated interest for low-loss nanophotonics, nonlinear optical phenomena, and microelectromechanical systems (MEMS). Recently, SiC has also shown promise as a host for optically addressable spin defects. These include the neutral divacancy (VV$^0$), the silicon vacancy (V$_{Si}$), and substitutional transition-metal ions (Cr$^{4+}$, V$^{4+}$, Mo$^{5+}$), among others. For these defects, isolated electronic states formed in the band gap create spin sublevels. The spin state can then be manipulated with applied microwave fields and read out using their distinct levels of photoluminescence (PL) after optical excitation. Experiments have utilized this optical readout mechanism to demonstrate control of the ground-state spin, forming the basis of a qubit. Additionally, the near-infrared emission of many of these SiC defects makes them compatible with existing fiber optic networks that operate at telecom wavelengths. For the V$_{Si}$ and VV$^0$, investigation of the excited-state optical fine structure has also revealed spin-preserving transitions that can be individually addressed in high-quality samples. The combination of ground-state spin control with optical spin readout using these transitions lays the foundation for a high-fidelity spin-to-photon interface, with potential applications in quantum communication, distributed quantum computing, and quantum sensing.

For point-defect qubits in semiconductors such as SiC, an overarching goal is the development of a long-distance interconnected quantum network, where electron spins act as stationary qubit nodes interconnected by single photons acting as carriers of quantum information. This architecture could then be utilized as a “quantum repeater” to relay quantum states over length scales beyond the $\sim 100$ km limit of single photons through fiber. However, entanglement rates and scalability are limited by intrinsic emission into the zero-phonon line (ZPL), which is used to produce indistinguishable photons for interference between spatially separated spins. To this end, the defect spin community has explored using photonic nanocavities to enhance a coupled defect’s ZPL emission. This enhancement is typically expressed as the Purcell factor.
Purcell factor, which quantifies an excited state’s lifetime reduction as a ratio of emission rates:

\[
F = \frac{\Gamma_{\text{cavity}}}{\Gamma_{\text{bulk}}} = F_1 \cdot F_2 \cdot \frac{3Q}{4\pi^2 V} \left( \frac{\lambda_{\text{cavity}}}{n} \right)^3 + 1
\]

(1)

where \(\Gamma_{\text{cavity}}\) and \(\Gamma_{\text{bulk}}\) are the cavity-enhanced and unmodified emission rates, with \(F = 1\) defining no enhancement. For the photonic cavity, \(Q\) is the quality factor, \(V\) is the mode volume, \(\lambda_{\text{cavity}}\) is the resonant wavelength, and \(n\) is the index of refraction. The terms \(F_1\) and \(F_2\) represent spatial overlap and spectral matching between the emitter and cavity mode, respectively, and are both equal to 1 in the case of perfect coupling (see Supporting Information). In recent work, cavity-defect systems in both diamond and silicon carbide have featured photonic crystal cavities with high quality factors (~\(10^5\)–\(10^6\)) and small mode volumes (~\(\lambda^3/n^3\)). For silicon carbide, in particular, its high-Q nanophotonic capabilities, intrinsic spin-defect emitters, and wafer-scale doping control situate it to be a highly promising platform for integrated spin-photonic systems. However, despite SiC’s potential, photonic integration with single VV0s has not been achieved.

In this Communication, we fabricate nanobeam photonic crystal cavities in 4H-SiC and couple them to single VV0s. We start with a description of the photonic cavity design and fabrication process. We then characterize a single VV0 within the cavity structure at cryogenic temperatures. When the cavity is tuned into resonance with the VV0, we observe a Purcell enhancement of ~50 and an improvement of the Debye–Waller (DW) factor from ~5% to ~70–75%. Lastly, we demonstrate microwave control of the ground-state spin and measure spin coherence times. This union between single defect control and cavity-emitter interactions results in significant increases in the VV0’s ZPL emission with coherent electron spin states, establishing important groundwork for single-shot readout and scalable remote spin entanglement using defect spins.

**Cavity Fabrication and Characterization.** To create a photonic nanocavity, light must be confined in all three dimensions. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work. Archetypal structures employ a submicron thin slab of dielectric material to provide out-of-plane confinement through total internal reflection and a patterning of Bragg mirrors to provide in-plane confinement. This results in either a one-dimensional (1D) or two-dimensional (2D) photonic crystal design, with both systems demonstrating high quality factors with small mode volumes (~\(\lambda^3/n^3\)). For this work, we select a 1D nanobeam structure due to its more compact size and successful demonstration in previous work.
To form the nanobeam cavities, we utilize electron beam lithography for in-plane patterning and photoelectrochemical (PEC) etching for creating an undercut structure. The fabrication procedure, outlined in Figure 1b, begins with electron beam lithography to define a thin nickel mask with evaporation and liftoff. Next, a SF$_6$-based inductively coupled plasma (ICP) etches through the silicon carbide in the regions not protected by the nickel. After an acid clean to remove the metal, a PEC etch and subsequent HF clean selectively etches the layer of p-type 4H-SiC $400 \text{ nm}$ below the top surface, suspending the nanobeams. A scanning electron microscopy (SEM) image of a representative device is shown in Figure 1c. The nanobeams appear smooth both on the topside and sidewalls of the beams, with relatively smooth and vertical etched holes. We employed a variety of cavity dimensions to create cavity resonances that include ZPLs for each of the $\sigma^{(\text{hh})}$, $\sigma^{(\text{kk})}$, and $\sigma^{(\text{kh})}$ transitions (see Supporting Information for nomenclature).

Single $\sigma^{(\text{V})}$ Characterization. After creating defects with an electron irradiation procedure (see Supporting Information), we characterize a single $\sigma^{(\text{V})}$ coupled to the cavity in Figure 1d. Figure 2a shows a spatial PL scan taken at 5 K with off-resonant (905 nm) excitation and the cavity off/on resonance with an $\sim1078 \text{ nm}$ $\sigma^{(\text{V})}$ transition. Subsequent photoluminescence excitation (PLE) measurements reveal two peaks at frequencies of 277.984 and 278.027 THz (Figure 2b). We then perform pulsed optically detected magnetic resonance (ODMR) with resonant optical excitation and a nearby wire-bond to drive microwave spin transitions. This results in an ODMR peak centered at 1.328 GHz (Figure 2c, center), which is closest to the $\sigma^{(\text{hh})}$ $\sigma^{(\text{V})}$ transition at 1.336 GHz.

As we vary the strength of an applied $c$-axis oriented magnetic field, this resonance separates into two lines due to a Zeeman splitting. The observed shifts at $\sim2.76 \text{ MHz/G}$ match closely with the electron gyromagnetic ratio of 2.8 MHz/G found in the $c$-axis ($\sigma^{(\text{hh})}$) defects. The presence of only one ODMR peak under zero magnetic field indicates that the transverse zero-field splitting (E) is approximately zero in the $\sigma^{(\text{V})}$ spin Hamiltonian, which is also consistent with a $c$-axis oriented $\sigma^{(\text{V})}$. If we instead apply off-resonant optical excitation, we observe ODMR with a negative contrast that matches previous work with $\sigma^{(\text{hh})}$ $\sigma^{(\text{V})}$s (see Supporting Information). Thus, while the ZPL of this defect matches the $\sigma^{(\text{hh})}$ $\sigma^{(\text{V})}$ wavelength ($\sim1078 \text{ nm}$), the $c$-axis spin orientation and the off-resonant ODMR contrast sign indicate the presence of an $\sigma^{(\text{hh})}$ $\sigma^{(\text{V})}$. We attribute this behavior to a highly strained environment (see Supporting Information), likely due to the high doping levels used during growth.
Additionally, we confirm the presence of a single optical emitter with a second-order correlation measurement under resonant excitation (Figure 2d). The antibunching dip \( g^{(2)}(0) \leq 0.5 \) indicates the presence of a single emitter, and the value \( g^{(2)}(0) = 0.096 \) indicates that this \( \text{VV}^0 \) is an excellent single-phonon source. Meanwhile, the observed bunching behavior is indicative of nonradiative transitions from the excited state. Solving the rate equations for this system (see Supporting Information) and fitting it to the observed \( g^{(2)} \) gives an effective dark state lifetime of \( \tau_{\text{dark}} \approx 60.7 \) ns. The nonradiative transitions are likely a combination of intersystem crossing (ISC) decays and \( \text{VV}^0 \) ionization. Although the ISC rates have not been explored in 4H-SiC \( \text{VV}^0 \)'s, in the 3C \( \text{VV}^0 \) they were estimated to be on a similar time scale of \( \approx 50-100 \) ns.\(^17\) Additionally, \( \text{VV}^0 \) ionization can be observed in our experiment under lower laser powers as a blinking behavior. Without a sufficiently strong 905 nm charge reset pulse, the \( \text{VV}^0 \) may be trapped in a nonradiative charge state for long periods of time, as has been observed in other work.\(^42,43\)

**Purcell Enhancement.** With a tunable photonic nanocavity and a \( \text{VV}^0 \) emitter within its mode volume, we are able to observe Purcell enhancement of the \( \text{VV}^0 \)'s optical emission. When the cavity is off resonance with the \( \text{VV}^0 \) and addressed with an off-resonant 905 nm laser, two peaks at \( \approx 1078 \) nm can be observed in a PL spectrum (Figure 3a, top inset). These peak locations and their \( \approx 40 \) GHz splitting correspond to thePLE peaks observed under resonant excitation (Figure 2b). We will label the lower/higher energy transitions as the lower/upper branches of the orbital fine structure, respectively.\(^17\) When the cavity is then tuned into resonance with the defect, a significant increase in emission is observed, with selective enhancement of the lower branch shown in Figure 3a. This count rate increase correlates closely with the Purcell factor, which in this case is given by

\[
F = \frac{I_{\text{ZPL, on}}}{I_{\text{ZPL, off}}}
\]  

(2)

where \( I_{\text{ZPL, on}} \) and \( I_{\text{ZPL, off}} \) represent the ZPL intensity when the cavity is on resonance and blueshifted off resonance, respectively. This equation matches the form of eq 1, with ZPL intensities acting as measures of emission rates. Integrating the counts under the two peaks when off and on cavity resonance gives Purcell factors of \( \approx 53 \) (Figure 3a) and \( \approx 16 \) (see Supporting Information) for the lower and upper branches, respectively. This difference could be explained by differing optical dipole orientations of the two branches, which would give varied matching to the cavity mode. A similar effect was observed for cavity enhancement of \( \text{V}_{\text{Si}} \) defects in silicon carbide, which also displays two rotated optical dipoles.\(^27\)

To corroborate the presence of Purcell enhancement, we directly measured excited-state lifetimes with the cavity on and off resonance with the \( \text{VV}^0 \). Using resonant excitation pulses from an electro-optic modulator, we observe an off-resonance lifetime of \( \tau_{\text{off}} = 15.7 \pm 0.3 \) ns (consistent with bulk measurements\(^17\)) and an on-resonance lifetime of \( \tau_{\text{on}} = 5.3 \pm 0.1 \) ns (Figure 3b). The relationship between measurable lifetimes and the Purcell factor is given by

\[
F = \frac{\tau_{\text{dark}}(\tau_{\text{off}} - \tau_{\text{on}})}{\alpha(\tau_{\text{dark}} - \tau_{\text{off}})} + 1
\]

(3)

where, for the \( \text{VV}^0 \), \( \tau_{\text{dark}} \) is the combined lifetime of all nonradiative decays, \( \tau_{\text{off}} \) is the lifetime off cavity resonance, \( \tau_{\text{on}} \) is the lifetime on cavity resonance, and \( \alpha \) is the intrinsic DW factor (see Supporting Information). Combining our measurements with a previously measured \( \approx 5.3\% \) DW factor\(^\beta\) gives a Purcell factor of \( F \approx 51 \), which is in good agreement with the value of \( F \approx 53 \) from spectral measurements.

One of the direct consequences of a Purcell enhancement is an increased Debye–Waller factor, which follows the relation

\[
F = \frac{\beta(\alpha - 1)}{\alpha(\beta - 1)}
\]

(4)

where \( \alpha \) and \( \beta \) represent the \( \text{VV}^0 \)'s DW factor off and on cavity resonance, respectively (see Supporting Information). For our sample, spatially varying background luminescence from nitrogen vacancy (NV) centers in the n-doped silicon carbide\(^5,7,16,44\) makes it difficult to directly integrate spectrometer counts to obtain \( \alpha \) and \( \beta \). However, we do observe background-subtracted count rates of 120 and 460 kCts/s when off and on cavity resonance, which allows us to estimate an on-resonance DW factor of \( \beta \approx 75\% \) and a Purcell factor of \( F \approx 54 \) (see Supporting Information). These numbers match well with the Purcell factors of \( \approx 53 \) and \( \approx 51 \) obtained from Figure 3 and the corresponding 74% DW factor from eq

---

**Figure 3.** Purcell enhancement of a single \( \text{VV}^0 \) in a photonic crystal cavity. (a) Emission spectrum of the \( \text{VV}^0 \) when excited with off-resonant 905 nm laser light with the cavity on (inset, lower right) and off (inset, upper right) resonance with the lower energy branch. A ratio of emission intensities gives a Purcell factor of \( \approx 53 \). The on-resonance trace for the combined plot is vertically offset for clarity. (b) Lifetime measurements of the \( \text{VV}^0 \) under resonant 277.984 THz excitation with the cavity on and off resonance. Fits to an exponential decay of \( \exp(-t/\tau) \) give a shortened lifetime \( (\tau = 5.3 \pm 0.1 \) ns\) with 95% confidence intervals, giving a Purcell factor of \( \approx 51 \). All measurements were taken at 5 K.
We then apply Ramsey interferometry and Hahn echo pulse sequences on the same |0⟩ → |±1⟩ transition to determine the spin dephasing and spin coherence times. Under a c-axis magnetic field of ~218 G, we obtain a dephasing time of $T_\phi^0 = 592 \pm 18$ ns (Figure 4b) and a decoherence time of $T_2 = 9.3 \pm 2.0$ μs (Figure 4c). Under a lower magnetic field of ~6 G, we obtain similar times of $T_\phi^0 = 605 \pm 33$ ns and $T_2 = 7.4 \pm 0.6$ μs (see Supporting Information), indicating that coherence in this sample is not primarily limited by the SiC nuclear spin bath.

Collectively these times are shorter than previous reports of $T_\phi^0 \approx 1–2$ μs and $T_2 \approx 1.2$ ms in bulk SiC c-axis VV0s, with the discrepancy likely arising from magnetic dipole interactions with electron spins from n-type dopants and surface charge traps.57,48 It is worth noting that, for a VV0 located in an unfabricated NIN epilayer, we measure an improved $T_\phi^0 = 4.01 \pm 0.38$ μs and $T_2 = 200 \pm 27.6$ μs under ~218 G (see Supporting Information). Therefore, it appears the fabrication process introduces additional decoherence sources, potentially from increased surfaces or crystal damage. However, there is a variety of approaches to offset these effects. The PEC undercut could likely be performed at lower doping levels, and postfabrication surface treatments could potentially be used to limit the presence of surface charge traps.45,50

In the regime where $T_1$ is significantly longer than $T_2$, it is possible to extend spin coherence through dynamical decoupling sequences. For the cavity VV0, in two separate
Discussions

Experimental increases in both the Debye–Waller factor and PL count rate have significant implications for enhancing the entanglement generation rate between VV0 spins. In the Barrett-Kok protocol, for example, the entanglement success rate is proportional to the square of the DW factor for two consecutive ZPL detection events. Using the ~75% DW factor measured here then gives a significant projected entanglement rate increase of ~200 between two (hh) VV0s. Additionally, entanglement verification relies on single-shot readout to determine the electron spin state in a single measurement, which is ultimately limited by photon detection throughput. For this system, the Purcell-enhanced threefold lifetime decrease would correspond to triple the emission events before a spin flip. The overall increase of off-cavity-resonance PL (~120 kCts/s) compared to bulk VV0s (typically ~20–50 kCts/s) is also indicative of a slightly improved collection efficiency. This is a vital factor for single-shot readout measurements, since a majority of PL from bulk VV0s is lost due to refraction at the SiC/air interface. Thus, both single-shot readout of VV0 spin states in a photonic structure and photonic enhanced entanglement could be achievable in future studies.

The cavity-enhanced VV0 studied here contains 4–5 GHz optical line widths comparable to those seen in near surface NV centers in diamond, but above the lifetime limit of ~11 MHz. We attribute the broadened optical line widths to spectral diffusion originating from a fluctuating charge environment around the defect. These charge fluctuations could be from nearby doped regions, other nearby defects and impurities, or surface charge traps. It is worth noting that the optical line widths are broader in 400 nm suspended-type SiC (~90–20 GHz) compared to the suspended NIN shown here (~4–5 GHz). Thus, doping configurations and growth conditions can have significant effects on spectral diffusion, opening the possibility to achieve narrow line widths through properly doped structures. Additionally, optical line widths are ~1 GHz for defects in the intrinsic layer of NINPM material before fabrication (see Supporting Information), indicating that the fabrication process or final nanostructure is a significant source of broadening. To counteract this effect, surface treatments or applied voltages could be used to maintain narrow line widths. Under applied electric fields, for example, VV0 optical line widths as narrow as ~20 MHz have been observed.

In conclusion, we have fabricated a photonic crystal cavity in silicon carbide coupled to a single VV0. We observe Purcell enhancement of the ZPL optical transition with a Purcell factor of ~50, a subsequent increase in Debye–Waller factor from ~5% to ~70–75%, and coherent spin control of the VV0 ground state with coherence extension. The use of a doped nanostructure allows for potential electric field and charge control, in situ Stark tuning, and improved collection efficiencies for optimized geometries, all of which would provide further improvements to the VV0 optical properties. As a whole, this system advances the robustness of spin-to-photon transduction for the VV0 in a technologically mature material. Looking beyond to many-qubit architectures, photonic nanocavities will be a necessary component to maintain scalability across long-distance entanglement networks.

Associated Content

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c00339.

Details on VV0 creation procedure, SiC doping, fabrication details, VV0 nomenclature conventions, and cavity tuning procedure. Additional cavity VV0 spin coherence measurements and measurements with off-resonant optical excitation. Characterization of bulk defects in the NINPM material. Observation of Purcell enhancement of upper branch of the cavity VV0. Discussion of modeled cavity mode with relevant dimensions. Details on photoelectrochemical etching. Details on rate equations used to model the system. Derivations of Purcell factor expressions. Discussion of the effect of strain on the cavity VV0 (PDF)

Author Information

Corresponding Author

David D. Awschalom — Pritzker School of Molecular Engineering and Department of Physics, University of Chicago, Chicago, Illinois 60637, United States; Center for Molecular Engineering and Materials Science Division, Argonne National Laboratory, Lemont, Illinois 60439, United States; orcid.org/0000-0002-8591-2687; Email: awsch@uchicago.edu

Authors

Alexander L. Crook — Pritzker School of Molecular Engineering and Department of Physics, University of Chicago, Chicago, Illinois 60637, United States
Christopher P. Anderson — Pritzker School of Molecular Engineering and Department of Physics, University of Chicago, Chicago, Illinois 60637, United States
Kevin C. Miao — Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States
Alexandre Bourassa — Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States
Hope Lee — Pritzker School of Molecular Engineering and Department of Physics, University of Chicago, Chicago, Illinois 60637, United States
Sam L. Bayliss — Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States; orcid.org/0000-0002-1156-7243
David O. Bracher — John A. Paulson School of Engineering and Applied Sciences and Department of Physics, Harvard University, Cambridge, Massachusetts 02138, United States
Xingyu Zhang — John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, United States
Hirosi Abe — National Institutes for Quantum and Radiological Science and Technology, Takasaki, Gunma 370-1292, Japan

Takeshi Ohshima — National Institutes for Quantum and Radiological Science and Technology, Takasaki, Gunma 370-1292, Japan; orcid.org/0000-0002-7850-3164

Evelyn L. Hu — John A. Paulson School of Engineering and Applied Sciences and Department of Physics, Harvard University, Cambridge, Massachusetts 02138, United States

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.nanolett.0c00339

Author Contributions
A.C. developed, fabricated, and measured the photonic crystal devices. C.A. and H.L. aided with fabrication procedures. C.A., K.M., and A.B. aided with optical characterization, cryogenic spin measurements, and analysis of data. S.B. assisted with resonant lifetime measurements and development of the three-level $g^{(2)}$ model. D.B., X.Z., and E.H. were instrumental in the development of both the PEC etch of SiC and SiC photonic devices. H.A. and T.O. performed electron irradiation of SiC samples to create divacancies. D.A. oversaw and directed the project. A.C., C.A., K.M., A.B., H.L., and S.B. all contributed with the drafting of the manuscript.

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS
This work was supported by the NSF EFRI AQUIRE EFMA-1641099 and the Univ. of Chicago MRSEC DMR-1420709. (A.C.) This work was funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences. This work also made use of the Pritzker Nanofabrication Facility part of the Pritzker School of Molecular Engineering at the Univ. of Chicago, which receives support from Soft and Hybrid Nanotechnology Experimental (SHyNE) Resource (NSF ECCS-1542205), a node of the National Science Foundation’s National Nanotechnology Coordinated Infrastructure. Electron irradiation was funded by Grant Nos. JSPS KAKENHI 17H01056 and 18H03770. This work was completed in part with resources provided by the Univ. of Chicago’s Research Computing Center.

REFERENCES
(1) Castelletto, S.; Rosa, L.; Johnson, B. C. Silicon Carbide for Novel Quantum Technology Devices. In Advanced Silicon Carbide Devices and Processing; Intech Open, 2015; pp 222–248.
(2) Guo, X.; Xun, Q.; Li, Z.; Du, S. Silicon carbide converters and MEMS devices for high-temperature power electronics: A critical review. Micromachines 2019, 10, 406.
(3) Zorman, C. A.; Parro, R. J. Micro- and nanomechanical structures for silicon carbide MEMS and NEMS. Phys. Status Solidi B 2008, 245, 1404–1424.
(4) Koehl, W. F.; Buckley, B. B.; Heremans, F. J.; Calusine, G.; Awschalom, D. D. Room temperature coherent control of defect spin qubits in silicon carbide. Nature 2011, 479, 84–87.
(5) Christie, D. J.; et al. Isolated electron spins in silicon carbide with millisecond coherence times. Nat. Mater. 2015, 14, 160–163.
(6) Csőre, A.; Von Bardeleben, H. J.; Cantin, J. L.; Gali, A. Characterization and formation of NV centers in 3C, 4H, and 6H SiC: An ab initio study. Phys. Rev. B: Condens. Matter Mater. Phys. 2017, 96, 085204.
(7) Wang, J. F.; et al. Coherent control of nitrogen-vacancy center spins in silicon carbide at room temperature. arXiv, 1909.12481 (2019). https://arxiv.org/abs/1909.12481 (accessed 2020-03-24).
(8) Lohmann, A.; Johnson, B. C.; McCallum, J. C.; Castelletto, S. A review on single photon sources in silicon carbide. Rep. Prog. Phys. 2017, 80, 034502.
(9) Widmann, M.; et al. Coherent control of single spins in silicon carbide at room temperature. Nat. Mater. 2015, 14, 164–168.
(10) Nagy, R.; et al. Quantum Properties of Dichroic Silicon Vacancies in Silicon Carbide. Phys. Rev. Appl. 2018, 9, 25–27.
(11) Nagy, R.; et al. High-fidelity spin and optical control of single silicon-vacancy centres in silicon carbide. Nat. Commun. 2019, 10, 1954.
(12) Wólfowicz, G.; et al. Vanadium spin qubits as telecom quantum emitters in silicon carbide. arXiv, 1908.09817 (2019). https://arxiv.org/abs/1908.09817 (accessed 2020-03-24).
(13) Diler, B.; et al. Coherent control and high-fidelity readout of chromium ions in commercial silicon carbide. npj Quantum Inf 2020, 6, 11.
(14) Koehl, W. F.; et al. Resonant optical spectroscopy and coherent control of Cr$^4+$ spin ensembles in SiC and GaN. Phys. Rev. B: Condens. Matter Mater. Phys. 2017, 95, 035207.
(15) Bosma, T.; et al. Identification and tunable optical coherent control of transition-metal spins in silicon carbide. npj Quantum Inf 2018, 4, 48.
(16) Von Bardeleben, H. J.; et al. NV centers in 3C,4H, and 6H silicon carbide: A variable platform for solid-state qubits and nanosensors. Phys. Rev. B: Condens. Matter Mater. Phys. 2016, 94, 121102.
(17) Christie, D. J.; et al. Isolated spin qubits in SiC with a high-fidelity infrared spin-to-photon interface. Phys. Rev. X 2017, 7, 021046.
(18) Banks, H. B.; et al. Resonant Optical Spin Initialization and Readout of Single Silicon Vacancies in 4H - Si C. Phys. Rev. Appl. 2019, 11, 024013.
(19) Wehner, S.; Elkouss, D.; Hanson, R. Quantum internet: A vision for the road ahead. Science 2018, 362, No. eaam9288.
(20) Briegel, H. J.; Dür, W.; Cirac, J. I.; Zoller, P. Quantum repeaters: The role of imperfect local operations in quantum communication. Phys. Rev. Lett. 1998, 81, 5932–5935.
(21) Childress, L.; Taylor, J. M.; Sørensen, A. S.; Lukin, M. D. Fault-tolerant quantum communication based on solid-state photon emitters. Phys. Rev. Lett. 2006, 96, 96–99.
(22) Pelton, M. Modified spontaneous emission in nanophotonic structures. Nat. Photonics 2015, 9, 427–435.
(23) Li, L.; et al. Coherent spin control of a nanocavity-enhanced qubit in diamond. Nat. Commun. 2015, 6, 6173.
(24) Calusine, G.; Politi, A.; Awschalom, D. D. Silicon carbide photonic crystal cavities with integrated color centers. Appl. Phys. Lett. 2014, 105, 011123.
(25) Calusine, G.; Politi, A.; Awschalom, D. D. Cavity-Enhanced Measurements of Defect Spins in Silicon Carbide. Phys. Rev. Appl. 2016, 6, 014019.
(26) Bracher, D. O.; Hu, E. L. Fabrication of High-Q Nanobeam Photonic Crystals in Epitaxially Grown 4H-SiC. Nano Lett. 2015, 15, 6202–6207.
(27) Bracher, D. O.; Zhang, X.; Hu, E. L. Selective Purcell enhancement of two closely linked zero-phonon transitions of a silicon carbide color center. Proc. Natl. Acad. Sci. U. S. A. 2017, 114, 4060–4065.
(28) Lukin, D. M.; Dory, C.; Mishra, S. D.; Guidry, M. A.; Radulaski, M.; et al. 4H-Silicon-Carbide-on-Insulator for Integrated Quantum and Nonlinear Photonics. Nat. Photonics 2019, DOI: 10.1038/s41566-019-0556-6.
(29) Hausmann, B. J. M.; et al. Coupling of NV centers to photonic crystal nanobeams in diamond. Nano Lett. 2013, 13, 5791–5796.
(30) Lee, J. C.; et al. Deterministic coupling of delta-doped nitrogen vacancy centers to a nanobeam photonic crystal cavity. Appl. Phys. Lett. 2014, 105, 261101.
(31) Song, B.-S.; et al. Ultrahigh-Q photonic crystal nanocavities based on 4H silicon carbide. *Optica* 2019, 6, 991–995.

(32) Lee, J. Y.; Lu, X.; Lin, Q. High-Q silicon carbide photonic-crystal cavities. *Appl. Phys. Lett.* 2015, 106, 041106.

(33) Deotare, P. B.; McCutcheon, M. W.; Frank, I. W.; Khan, M.; Lončar, M. High quality factor photonic crystal nanobeam cavities. *Appl. Phys. Lett.* 2009, 94, 121106.

(34) Sekoguchi, H.; Takahashi, Y.; Asano, T.; Noda, S. Photonic crystal nanocavity with a Q-factor of ~ 9 million. *Opt. Express* 2014, 22, 916–924.

(35) Zhao, F.; Islam, M. M.; Huang, C. F. Photoelectrochemical etching to fabricate single-crystal SiC MEMS for harsh environments. *Mater. Lett.* 2011, 65, 409–412.

(36) Pavvuny, S. P.; et al. On the doping concentration dependence and dopant selectivity of photogenerated carrier assisted etching of 4H–SiC epilayers. *Electrochim. Acta* 2019, 323, 134778.

(37) Falk, A. L.; et al. Polytype control of spin qubits in silicon carbide. *Nat. Commun.* 2013, 4, 1819.

(38) Huh, S. W.; et al. Doping-induced strain and relaxation of Al-doped 4H-SiC homoepitaxial layers. *J. Appl. Phys.* 2004, 96, 4637–4641.

(39) Jacobson, H.; et al. Doping-induced strain in N-doped 4H-SiC crystals. *Appl. Phys. Lett.* 2003, 82, 3689–3691.

(40) Chung, H. J.; Liu, J. Q.; Henry, A.; Skowronski, M. Stacking Fault Formation in Highly Doped 4H-SiC Epilayers during Annealing. *Mater. Sci. Forum* 2003, 433–436, 253–256.

(41) Okojie, R. S.; Holzheu, T.; Huang, X. R.; Dudley, M. X-ray diffraction measurement of doping induced lattice mismatch in n-type 4H-SiC epilayers grown on p-type substrates. *Appl. Phys. Lett.* 2003, 83, 1971–1973.

(42) Anderson, C. P.; et al. Electrical and optical control of single spins integrated in scalable semiconductor devices. *Science* 2019, 366, 1225–1230.

(43) Wólfowicz, G.; et al. Optical charge state control of spin defects in 4H-SiC. *Nat. Commun.* 2017, 8, 1876.

(44) Zargaleh, S. A.; et al. Nitrogen vacancy center in cubic silicon carbide: A promising qubit in the 1.5 μm spectral range for photonic quantum networks. *Phys. Rev. B: Condens. Matter Mater. Phys.* 2018, 98, 165203.

(45) Miao, K. C.; et al. Electrically driven optical interferometry with spins in silicon carbide. *Sci. Adv.* 2019, 5, No. eaay0527.

(46) Seo, H.; et al. Quantum decoherence dynamics of divacancy spins in silicon carbide. *Nat. Commun.* 2016, 7, 12935.

(47) Rosskopf, T.; et al. Investigation of surface magnetic noise by shallow spins in diamond. *Phys. Rev. Lett.* 2014, 112, 147602.

(48) Barry, J. F.; et al. Sensitivity Optimization for NV-Diamond Magnetometry *arXiv*, 1903.08176 (2019). https://arxiv.org/abs/1903.08176 (accessed 2020-03-24).

(49) Kim, M.; et al. Decoherence of Near-Surface Nitrogen-Vacancy Centers Due to Electric Field Noise. *Phys. Rev. Lett.* 2015, 115, 087602.

(50) Fávaro De Oliveira, F.; et al. Effect of low-damage inductively coupled plasma on shallow nitrogen-vacancy centers in diamond. *Appl. Phys. Lett.* 2015, 107, 073107.

(51) Meiboom, S.; Gill, D. Modified spin-echo method for measuring nuclear relaxation times. *Rev. Sci. Instrum.* 1958, 29, 688–691.

(52) Barrett, S. D.; Kok, P. Efficient high-fidelity quantum computation using matter qubits and linear optics. *Phys. Rev. A: At., Mol., Opt. Phys.* 2005, 71, 060310.

(53) Bernien, H.; et al. Heralded entanglement between solid-state qubits separated by three metres. *Nature* 2013, 497, 86–90.

(54) Robledo, L.; et al. High-fidelity projective read-out of a solid-state spin quantum register. *Nature* 2011, 477, 574–578.

(55) Faraon, A.; Santori, C.; Huang, Z.; Acosta, V. M.; Beausoleil, R. G. Coupling of nitrogen-vacancy centers to photonic crystal cavities in monocrystalline diamond. *Phys. Rev. Lett.* 2012, 109, 033604.

(56) Liu, J.; et al. Single Self-Assembled InAs/GaAs Quantum Dots in Photonic Nanostructures: The Role of Nanofabrication. *Phys. Rev. Appl.* 2018, 9, 064019.