Laser writing of spin defects in nanophotonic cavities

High-yield engineering and characterization of cavity–emitter coupling is an outstanding challenge in developing scalable quantum network nodes. Ex situ defect formation systems prevent real-time analysis, and previous in situ methods are limited to bulk substrates or require further processing to improve the emitter properties. Here we demonstrate the direct laser writing of cavity-integrated spin defects using a nanosecond pulsed above-bandgap laser. Photonic crystal cavities in 4H-silicon carbide serve as a nanoscope monitoring silicon-monovacancy defect formation within the approximately 200 nm$^3$ cavity-mode volume. We observe spin resonance, cavity-integrated photoluminescence and excited-state lifetimes consistent with conventional defect formation methods, without the need for post-irradiation thermal annealing. We further find an exponential reduction in excited-state lifetime at fluences approaching the cavity amorphization threshold and show the single-shot annealing of intrinsic background defects at silicon-monovacancy formation sites. This real-time in situ method of localized defect formation, paired with cavity-integrated defect spins, is necessary towards engineering cavity–emitter coupling for quantum networking.

Optically active solid-state spin defects are a favourable candidate for quantum networking due to their spin–photon interface, long spin coherence, coupling to nuclear spin memories and natural integration into photonic crystal cavities (PCCs). PCCs augment the optical properties of defects via the Purcell effect by enhancing the emission of zero-phonon-line (ZPL) photons. Nanophotonic cavities additionally enable robust spin–photon entanglement interfaces that form the basis of cavity quantum electrodynamics protocols. Defect placement is central to engineering large cavity–emitter cooperativity for the optimal implementation of these proposals. Conventionally, ex situ focused ion beam and masked ion implantation have been used to locally incorporate solid-state spin defects. Alternatively, direct laser writing has been recently explored with nitrogen-vacancy centres in diamond, silicon monovacancy in 4H-silicon carbide (SiC) and neutral divacancy in 4H-SiC. Prior demonstrations of the laser writing of spin defects in diamond and 4H-SiC have utilized below-bandgap femtosecond pulsed lasers to locally form defects in the lattice. These previous methods have achieved control over defect number and spatial distribution, but have been limited to bulk materials, as optical, spin and materials properties near the surfaces have presented a challenge for nanophotonic integration. 'Ultrashort' femtosecond below-bandgap irradiation induces multiphoton excitation to generate electrons and holes from a transparent material, transferring energy to the lattice to form defects. For a wide-bandgap semiconductor, carrier generation is not limited to nonlinear absorption and can instead be generated through the linear absorption of above-bandgap photons. Consequently, the laser here writes defects using above-bandgap nanosecond pulses, where efficient linear absorption results in lower instantaneous pulse powers for defect creation. Spin-active defects are directly written into nanophotonic structures whose modal volumes (approximately

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1. John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA. 2. Department of Physics, Harvard University, Cambridge, MA, USA. 3. These authors contributed equally: Aaron M. Day, Jonathan R. Dietz. e-mail: ehu@seas.harvard.edu

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200 nm) provide insights into the local defect environment that were not available with prior systems. We verify the defects’ optical and spin properties by characterizing their photoluminescence (PL) and optically detected magnetic resonance (ODMR). Additionally, we study the relative damage imparted to the crystal lattice during laser irradiation by measuring the relationship of formation fluence to emitter lifetime. Finally, we observe the single-shot laser annealing of intrinsic defects on laser irradiation. In total, our method realizes in situ spin-defect integration into nanophotonics—a necessary step towards the scalable implementation of quantum networks.

Furthermore, 4H-SiC is an attractive quantum networking material platform: it is a wide-bandgap, fabrication-amenable semiconductor commercially available at the wafer scale, which hosts two promising near-infrared spin defects—VV\textsuperscript{\textregistered} and \(V_\text{Si}^-\). Recent results showing single-shot spin readout, preserved single-emitter coherence in nanophotonics, coherent acoustic spin control, and quantum and nonlinear optics on insulator demonstrate rapid progress towards a competitive commercial quantum networking platform\textsuperscript{10,12,16,17}. The nanoscale PCCs studied in this work are fabricated in unimplanted 4H-SiC using an electron-beam lithography, dry-etch and photoelectrochemical etch process outlined in another work\textsuperscript{14}. Our cavities are designed to be resonant with the optical transitions of the defects of interest, providing high sensitivity to the formation and evolution of defects under various processing conditions. The PCCs are ~250 nm thick; therefore, PL measurements in photonic structures reveal coupling to both emissive intrinsic surface defects and defects generated via laser irradiation. Intrinsic surface defects known to emit single photons are postulated to be related to the surface oxide and are routinely observed in 4H-SiC with emission spanning the 600–1,000 nm range\textsuperscript{10,12,16–20}.

Individual pulses of a nanosecond ultraviolet (UV) (337.1 nm) laser are focused onto the cavities (Fig. 1a), forming silicon-monovacancy defects at the irradiation site (Fig. 1b). Our method enables the real-time in situ monitoring of defect formation within the cavity-mode volume (Fig. 1a, inset). The individual pulse fluence and total pulse number incident on the cavities are varied to determine the optimal irradiation conditions, which deliver laser energy greater than a minimum threshold required to observe a change in the optical signature but lower than energies that amorphize the cavity (Supplementary Figs. 7–11). For single-pulse irradiation, the optimum range spanned approximately 0.4–0.9 \(\mu\)J per pulse, where irradiation-created \(V_\text{Si}^-\) emission is observed to be localized to the cavity-mode volume and decorated by cavity modes following the localized incorporation of defects into previously unirradiated material with intensity localized at the cavity-mode site. The spectra are integrated for 5 s.

The insets show the 77 K PL of the cavity-mode volume immediately before (orange) and after (blue) irradiation with a single 0.6 \(\mu\)J UV pulse, measured under 780 nm excitation. Before irradiation, the intrinsic surface-state emission weakly couples to the cavity, enabling the observation of resonant modes. After irradiation with a single UV pulse, a clear h-site ZPL is observed with the same cavity modes still present, indicating \(V_\text{Si}^-\) formation within the cavity-mode volume, and the preservation of the photonic properties of the device on irradiation. The spectrum is integrated for 180 s. The 300 K PL spectrum is decorated with cavity modes following the localized incorporation of defects into previously unirradiated material with intensity localized at the cavity-mode site. The spectra are integrated for 5 s.

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We then study single-pulse cavity irradiation under vacuum at 77 K. In situ irradiation spectroscopy enables the real-time observation of defect ZPLs, and subsequent room-temperature ODMR confirms the characteristic spin resonance (Fig. 2). Under high fluence, we observe ensemble emission possessing both h- and k-site ZPLs (Supplementary Fig. 8), and observe at decreased irradiation fluence emission from either an h- or k-site ZPL. Bright h-site $V^-_{\text{Si}}$ emission is observed to be integrated in a cavity with a quality factor of $Q = 2,600$. The characteristic k-site spin resonance observed in an adjacent cavity irradiated with a single 0.95 $\mu$J pulse.

**Fig. 2 | Preservation of cavity-mode optical and spin signatures on irradiation with a single UV pulse.** a, The 77 K PL spectrum of the cavity-integrated h-site silicon monovacancy formed with a single 0.475 $\mu$J UV pulse, showing no observable k-site emission. The h-site ZPL is observed at 861 nm with cavity modes decorating the phonon sideband at approximately 895 and 948 nm. The spectrum is integrated for 300 s. b, Fit of the 948 nm cavity mode of a, where $Q = 2,600$. c, The 77 K PL spectrum of narrow V2 ZPL formed from a single 0.6 $\mu$J UV pulse, with no observable h-site emission. d, The spectrum is integrated for 60 s and measured from an irradiation site with highly localized emission, where the k-site ZPL is present only within the diffraction-limited spot. The measurement is performed with 80 $\mu$W, 780 nm light. e, Characteristic k-site spin resonance observed in an adjacent cavity irradiated with a single 0.95 $\mu$J pulse.

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below-bandgap laser writing that inefficiently creates highly energetic carriers that may contribute to an avalanche ionization process, our above-bandgap method more efficiently provides electron–hole pairs and with much lower instantaneous laser power. We can gain insights into the efficiency and how the incident laser fluence might be apportioned by studying the lifetimes of the generated \( V^-_{\text{Si}} \) and their correlation with irradiation conditions. A dose array is generated as a function of fluence across numerous unimplanted PCCs (Supplementary Figs. 10 and 11) using a single irradiation pulse in one set of cavities and multiple pulses in the other. The excited-state lifetimes of the emitters formed from single- and multiple-pulse irradiation are plotted in Fig. 3a and their decays and fits are summarized in Fig. 3b–e. Data taken from a neighbouring unirradiated cavity and from a reference-ion-implanted cavity are also plotted for comparison. The transient decay data (Fig. 3b–d) reveal a short decay process that is comparable for all the laser-irradiated samples, with a mean

![Graph showing transient decay data for laser-irradiated cavity-integrated silicon-monovacancy defects.](image)

Fig. 3 | Lifetime analysis of laser-irradiated cavity-integrated silicon-monovacancy defects. a, Lifetimes plotted as a function of fluence, where SS and MS denote single-shot (one UV pulse) and multishot (two to three UV pulses), respectively. Short (\( \tau_s \)) and long (\( \tau_l \)) decay components are observed, with a substantial \( \tau_l \) dependence on the formation fluence and pulse count. The horizontal bars depict the decay of an ion-implanted cavity-integrated sample used as a comparison metric (black), and an unimplanted unirradiated cavity adjacent to the ones studied throughout the work (grey). The decay times are extracted from the fits shown in b–e, where the error bars (grey-shaded region surrounding the black fit trace) depict two standard deviations (s.d.) from the fit. Supplementary Table 1 summarizes the calculated lifetime fit values and standard deviation. We do not observe the cavity modification of the emitter lifetime due to spectral detuning between the emitter and cavity in conjunction with collection filtering on the ZPL.
lifetime of 393 ps, insensitive to irradiation conditions. The short decay component could be related to intrinsic defects on the surface or inherent in the material. Indeed, the lifetime of 250 ps measured in unimplanted, unirradiated cavities (Fig. 3e, light brown) is consistent with the distribution of short decay processes measured in the laser-irradiated cavities. This short lived component is attributable to surface defects, consistent with Fig. 2, where PL exhibits background luminescence, and the spin contrast of 0.2% is diminished relative to the 0.3–0.5% typically found in thermally unannealed ion-implanted near-surface emitters. This cavity nanoscope illuminates the presence of a lifetime component attributable to intrinsic surface defects inherent in the material or introduced by cavity fabrication.

In contrast, the long decay components (Fig. 3b–d) corresponding to the excited-state lifetime of $V'_C$ are strongly dependent on the irradiation conditions, revealing an exponential reduction at increased fluence and a uniform reduction by irradiation with multiple pulses. Increased fluence and pulse count may favour non-radiative decay channels or create other proximal defects that quench the monovacancies’ fluorescence. Thus, from the trends evident in Fig. 3a, we conclude that beneath the amorphization threshold of the cavities, lattice damage induced as irradiation fluence and pulse count is increased, resulting in reduced emitter lifetime and linewidth broadening.

We have already shown that surface defects have an impact on the measured spin and optical characteristics of laser-written cavities—these defects could also play a mediating role in $V'_C$ formation. Characterizing these states’ evolution under laser irradiation is important in ultimately developing an accurate physical model of the laser-writing process. Six cavities possessing pre-existing intrinsic surface defects are irradiated with a single UV pulse (Fig. 4a, blue box). We then measure the PL in both irradiated and unirradiated regions using a 632.8 nm continuous-wave helium–neon laser to efficiently excite the intrinsic defects. An avalanche photodiode raster scan reveals the surface defect emission, which appears as bright regions along the unirradiated edges of the cavities (Fig. 4a).

Surprisingly, the intrinsic defect PL is dimmer in the irradiated region. Comparing the PL of the irradiated cavity region (Fig. 4b, red) with the unimplanted, unirradiated surface defect emission (Fig. 4b, black), we note an 80% reduction in surface defect emission and an emergent peak centred at around 665 nm, commonly observed in the emission of a carbon antisite vacancy pair ($C_0V_C$) associated with the 640–670 nm A-line ZPLs. $C_0V_C$ has a lower calculated formation energy (~6.0 eV) than $V'_C$ (~7.5 eV) (ref. 29). Thus, it is not surprising that laser irradiation conditions that produce $V'_C$ also give rise to the observed $C_0V_C$; one mechanism may be the formation of $C_0V_C$ pairs from pre-existing $C_0$ and $V_C$. Our observed surface defect reduction with commensurate $C_0V_C$ and $V'_C$ formation is strikingly consistent with prior work exploring thermal processes for material enhancement. Having demonstrated here that $V'_C$ formation in cavities yields the laser annealing of unwanted intrinsic surface defects, future work intends to explore process optimization, specifically targeting surface treatment.

Further explorations are needed to more fully understand the mechanisms of defect creation for both above- and below-bandgap laser-writing techniques. Is it energetic electrons, electron–phonon coupling leading to local heating or some other effect produced by UV irradiation that creates defects in the material? We can gain insights from the considerable literature exploring laser-induced damage in semiconductors, via melting and dielectric breakdown. For the latter case, breakdown mechanisms include both avalanche ionization and multiphoton absorption. Avalanche ionization results from the production of high-energy electrons that can subsequently promote valence electrons directly. The high density of excess electrons, in turn, increases the absorption of the incident laser energy into the material. In general, this requires carrier concentrations on the order of $10^{24}$ cm$^{-3}$. We estimate that our laser excitation should produce $10^{23}$ cm$^{-3}$ (Supplementary Section X), but carrier diffusion constants, electron–phonon interactions and cavity geometry must also be taken into account: carriers can diffuse to recombine with sidewalls and surface states, similarly depleting the peak density of free carriers that can interact with the incident laser beam. This carrier diffusion and recombination may be responsible for the efficient single-shot cavity annealing observed in Fig. 4a. Furthermore, recombination-enhanced defect reactions have been observed on the above-bandgap illumination of semiconductors, where the injected carriers can be captured by defect sites to mediate highly localized multiphonon emission. Similarly, the energies required to effect the diffusion of intrinsic defects is calculated to be lower than their formation energies. Thus, laser irradiation might drive defect reactions among pre-existing defects. Indeed, UV nanosecond pulsed lasers have been shown to cause point defect damage in a wide array of material systems, where existing defects seed the generation of additional defects.

We have shown the single-shot cavity integration of $V'_C$ using above-bandgap nanosecond pulsed direct laser writing. The process preserves the spin and optical properties of the defect and the cavity mode of the nanophotonic device, demonstrating the lifetimes of near-surface spins longer than those observed for defects formed in previous laser-writing demonstrations where the defects were created micrometres below the surface in the bulk material. Additionally, we
have developed an understanding of the process that drives defect formation by studying lifetime quenching at increased irradiation pulse repeats and fluence. Furthermore, single-shot laser annealing reveals the suppression of surface states with the formation of $C_\text{sp}^3V_C$ and $V_\text{Ir}$, suggesting that pre-existing defects may play a mediating role in the laser-writing process. We have signatures supporting the fact that our process forms single $V_\text{Ir}$ by the count rate, spatial confinement, and isolation of h-site and k-site ZPLs at varied irradiation sites. The unambiguous observation of single-photon correlations is inhibited by the presence of surface defects (Supplementary Figs. 3 and 4), which could potentially be mitigated in future work by the laser writing of cavities on purer material or utilizing surface treatment methods to reduce background defect fluorescence. Although the present lifetime and ODMR data suggest that our method forms emitters with comparable background defect fluorescence, the high-throughput means of generating emitters and greatly eases the challenge of generating defects ex situ followed by large-scale characterization. The demonstrated technique allows not only the selective formation of defects but also the rapid in situ characterization of defects that are formed in cavities, only requiring the inclusion of one additional laser in the confocal system with no need for post-irradiation annealing to improve the emitter characteristics. Just as prior below-bandgap femtosecond laser writing has been pioneered in numerous material systems, the method here is relatively material-agnostic and can be extended to other desirable quantum networking platforms such as diamond and silicon. This process realizes defect formation with the single-site addressing of nanophotonic cavity modes, offering a tool in engineering optimal cavity–emitter coupling for system development in quantum networking and cavity quantum electrodynamics.

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**Methods**

**Sample preparation**
The sample used in this study is an epitaxially grown 100–200–100 nm p–i–p–n stack of 4H-SiC (Norstel AB), comprising $10^{21}$ cm$^{-3}$ aluminium-doped p-type, intrinsic-type and $10^{19}$ cm$^{-3}$ nitrogen-doped n-type bulk. The PCCs are fabricated on the sample using electron-beam lithography and the reactive ion etching process given elsewhere\(^\text{23-24}\). The fabrication process is finished with 900 °C annealing in air followed by a dip in hydrofluoric acid. We suspect that the fabrication process implemented with these devices could contribute to the increased concentration of intrinsic surface defects observed throughout this work.

The ion-implanted cavity sample used as a reference in Fig. 3 had the same epitaxial conditions and cavity fabrication as described above, but was implanted with $1 \times 10^{12}$ Si ions with an energy of 70 keV at 7° off the c axis to form an ensemble of V$_{\text{Si}}$ at a depth of 134 nm given by stopping range calculations using SRIM (ref. 24). The PL spectra of characteristic V$_{\text{Si}}$ emission measured from this sample are provided in Supplementary Figs. 13 and 14.

**Defect formation via pulsed laser writing**
A Spectra-Physics nanosecond pulsed nitrogen laser is used to direct individual pulses of 4 ns, 337.1 nm laser light onto unimplanted fabricated PCCs. The pulse fluence is controlled with a variable neutral density attenuator. The pulses are focused onto the sample with an Olympus 40× 0.6 numerical aperture correction-collar objective. The sample is mounted on a copper cold finger in a continuous-flow Janis ST-500 cryostat, held under a vacuum at approximately 5 × 10$^{-6}$ torr. The experiment is first performed at 300 K, where on the normal observation of cavity integration, the experiment is repeated and validated at 77 K.

**Experiment statistics**
Over the course of this study, hundreds of cavities at varied irradiation conditions were investigated. A dose grid of 64 different laser-irradiated devices are particularly studied in this work—a subset of which are presented in the main text and Supplementary Information. The PL spectra of cavity-integrated V$_{\text{Si}}$ (devices for which the cavity modes are observed decorating V$_{\text{Si}}$ emission) are presented from four different devices. One cavity is presented for which a narrow k-site ZPL is observed and the cavity mode is not evident, and one device is presented for characteristic laser-irradiated damage PL. The lifetime is observed and the cavity mode is not evident, and one device is presented for which a narrow k-site ZPL is observed and validated at 77 K.

**Data availability**
The data that support the findings of this work are presented in the Letter and the Supplementary Information. Source data are provided with this paper.

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**Author contributions**
A.M.D., J.R.D. and E.L.H. designed the research. A.M.D. and J.R.D. performed all the measurements and data analysis, with assistance from M.S. and M.Y. All the authors contributed to preparing the manuscript.

**Competing interests**
The authors declare no competing interests.
Additional information
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Correspondence and requests for materials should be addressed to Evelyn L. Hu.

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