### Supplementary information Purcell enhancement of a single silicon carbide color center with coherent spin control

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### Supplementary note 1: VV<sup>0</sup> creation procedure

In order to create  $VV^0$ s in silicon carbide, it is necessary to create damage in the pristine lattice. To achieve this, we first send samples to the National Institutes for Quantum and Radiological Science and Technology in Takasaki, Japan. There the silicon carbide is electron irradiated with relativistic 2 MeV electrons that damage the lattice uniformly and create single vacancies. After subsequent annealing at ambient pressure with Argon gas at 850 °C for 30 minutes, the vacancies migrate and form stable  $VV^0$  complexes. This is similar to the NV center formation mechanism in diamond, where single carbon vacancies migrate until finding a substitutional nitrogen to form the NV center complex. At an electron radiation dose of  $10^{16}$  electrons per square centimeter, we expect on the order of one  $VV^0$  per nanobeam mode volume. For the sample outlined in this study, we performed electron irradiation and annealing before the nanobeam fabrication process in order to confirm the presence of  $VV^0$ s.

### Supplementary note 2: SiC doping

Wafers were ordered commercially from Norstel and epitaxially grown to contain different doping layers. The SiC growth substrate is N-type 4H-SiC (12-30 m $\Omega \cdot$ cm) with a 4 degree off-axis growth. The doping and thicknesses of the NINPN layers are as follows: 500 µm of standard N-type buffer (1e<sup>18</sup> cm<sup>-3</sup> N), 3 µm of P-type (1e<sup>18</sup> cm<sup>-3</sup> Al), 100 nm of N-type (1e<sup>18</sup> cm<sup>-3</sup> N), 200 nm of I-type (<1e<sup>15</sup> cm<sup>-3</sup> residual dopants), and 100 nm of N-type (1e<sup>18</sup> cm<sup>-3</sup> N). Ohmic contact is made uniformly on the back n-type surface using ~300 nm of NiCr (80/20) for PEC etching. This n-type contact is made ohmic using a Solaris 150 RTP rapid thermal annealer for a 5 minute anneal with an Ar flow under ambient pressure at 950 °C.

### **Supplementary note 3: Fabrication**

Electron beam lithography was performed with a Raith EBPG5200 E-Beam lithography system with PMMA 495K A4 as a positive resist. Nickel was evaporated with an Angstrom NexDep Thermal E-beam evaporator. A PlasmaTherm ICP system was used for plasma etching of the SiC. We used a mixture of SF<sub>6</sub> and Ar gases at 40 and 10 sccm, respectively, with a bias power of 90 W and an ICP run power of 500 W at a process pressure of 6 mTorr. The SiC etch rate is roughly 350-400 nm/min under these conditions. The sample was acid cleaned with a piranha solution of  $1:3 H_2O_2:H_2SO_4$  followed by a 1:1 mixture of HF:HNO<sub>3</sub>.

### Supplementary note 4: Cavity tuning

A cavity resonance tuning mechanism is necessary to achieve maximal coupling between a photonic cavity and an emitter. We are able to tune the cavity resonant wavelength through gas condensation from heating and cooling of the cryostat. Over time, the nanobeam slowly accumulates condensed gas particles, which has the effect of slowly redshifting the cavity resonance. When the sample is brought to a higher temperature of ~30-40K, condensation is released from the nanobeam and the cavity resonance blueshifts. When a separate cooling stage of the cryostat is heated while the sample stage is left cool, condensation onto the beam increases and the cavity resonance redshifts. Through this method we are able to tune the cavity into resonance with an intrinsic VV<sup>0</sup> within a ~5 nm range, after which it remains stable for ~30 minutes.

## Supplementary note 5: VV<sup>0</sup> nomenclature.

The divacancy in silicon carbide consists of a silicon vacancy adjacent to a carbon vacancy. For the 4H-SiC crystal, the divacancy can have four orientations labeled as (hh), (kk), (hk), and (kh). In previous SiC defect nomenclature, the terminology PL1, PL2, PL3, PL4 has also been used for these four orientations, respectively. The *h* and *k* refer to inequivalent lattice sites in 4H-SiC corresponding to a hexagonal (h) or quasi-cubic (k) local crystal structure. For divacancy nomenclature, the first letter denotes the local structure of the carbon vacancy and the second letter denotes the local structure of the silicon vacancy. So, for example, the (kh) divacancy (also termed PL4) has a carbon vacancy at a quasi-cubic *k* site and an adjacent silicon vacancy at a hexagonal *h* site. The 4H-SiC polytype is labeled as 4H due to its 4-layer stacking (ABCB) of SiC bilayers and its hexagonal (H) crystal structure. Here, the letters ABC refer to the relative orientations of the silicon-carbon bilayers oriented along the [0001] *c*-axis. A and B are related by a translation, while C rotates the bilayer orientation by 60 degrees. The (hh) and (kk) divacancies are oriented along the *c*-axis of the lattice, while the (hk) and (kh) divacancies are oriented along basal directions.



**Figure S1** | **4H-SiC lattice and VV<sup>0</sup> orientation.** Every silicon carbide polytype can be composed by the appropriate repeating sequence of atomic bilayers. A single bilayer is defined here as a plane containing a silicon and carbon atom connected by *c*-axis oriented bond. The A bilayer is a translation of the B bilayer with no rotation, while a C bilayer is a 60 degree rotation of either the A or B bilayer. Here this is denoted as "twist" and "no twist". 4H-SiC is then a hexagonal lattice that follows the general repeating sequence of ABCB. Equivalently, this can be rewritten as CBAB, BABC, or BCBA. Regardless of the notation, the B bilayer occurs every other bilayer. For divacancies, the first letter denotes the carbon vacancy lattice site and the second letter denotes the silicon vacancy lattice site. In the 4H-SiC lattice, the *h* (hexagonal) and *k* (quasi-cubic) lattice sites alternate between silicon-carbon bilayers. Additionally, there are four inequivalent divacancy orientations for 4H-SiC, as shown in the figure.

### Supplementary note 6: Bulk and nanobeam NINPN VV<sup>0</sup> additional measurements

For comparison to the results obtained for the nanobeam VV<sup>0</sup>, we also collected data for a VV<sup>0</sup> in the intrinsic layer of the same NINPN-doped chip. We observe similar ZPL and ODMR shifts, indicating a uniform strain. Other defects in unpatterned regions of the chip displayed this same behavior. The improved optical linewidth of ~1 GHz and longer coherence times of  $T_2^* \approx 4 \,\mu s$  and  $T_2 \approx 200 \,\mu s$  suggest that fabricated nanostructures can hinder defect properties, a result established from previous cavity-defect work. Additionally, the position of the VV<sup>0</sup> within the intrinsically doped epilayer could cause coherence time variations based on the defect's proximity to doped layers.

We also collected statistics for optical linewidths and center frequencies of "bulk"  $VV^0s$  in the NIN epilayer and  $VV^0s$  in suspended nanobeams, as given below. In general the unfabricated  $VV^0$  defects displayed narrower optical linewidths, indicating that fabrication plays a role in affecting this metric. All  $VV^0s$  displayed similar center frequencies, with a similar variance between bulk and nanobeam  $VV^0s$ .



Bulk (n = 16),  $\mu = 1.37 \text{ GHz}$ ,  $\sigma_{sample} = 0.86 \text{ GHz}$ , range =  $0.5 \rightarrow 3.5 \text{ GHz}$ Nanobeam (n = 10),  $\mu = 5.20 \text{ GHz}$ ,  $\sigma_{sample} = 2.50 \text{ GHz}$ , range =  $2 \rightarrow 10 \text{ GHz}$ 

Center frequencies:

Bulk (n = 16),	$\mu = 277.957 \ THz$ ,	$\sigma_{sample} = 39.52 \ GHz$ ,	range = $106 GHz$
Beam $(n = 10)$ ,	$\mu = 278.016 THz$ ,	$\sigma_{sample} = 34.03 \ GHz$ ,	range = $126 GHz$



**Figure S2** | **Characterization of a bulk NIN VV<sup>0</sup>. (a)** Photoluminescence excitation (PLE), with 278.0015 THZ detuning. Optical linewidth from a Lorentzian fit is  $825 \pm 66$  MHz with a 95% confidence interval (b) Optically detected magnetic resonance (ODMR) collected on the same defect with resonant optical excitation and a low magnetic field of ~1.2 G parallel to the *c*-axis. (c) Ramsey sequence collected at 218 G parallel to the *c*-axis with a  $T_2^*$  confidence interval of 95% (b) Hahn echo sequence collected at 218 G with a  $T_2$  confidence interval of 95%.

#### Supplementary note 7: Cavity defect coherence time measurements at low-field

For the nanobeam VV<sup>0</sup> under a low magnetic field of ~6 G, we obtain coherence times of  $T_2^* = 605 \pm 33 \text{ ns}$  and  $T_2 = 7.4 \pm 0.6 \,\mu s$ , which are comparable to the high-field (218 G) values of  $T_2^* = 592 \pm 18 \text{ ns}$  and  $9.3 \pm 2.0 \,\mu s$  featured in the main text. This suggests that the nuclear spin bath is not the main cause for reduced coherence times in a nanostructure, but rather noise induced by nearby fabricated surfaces or doped layers. Additionally, in two separate measurements we observe spin relaxation times of  $T_1 = 1.02 \pm 0.47$  ms and  $T_1 = 2.43 \pm 1.58$  ms. Given the longer time scales for spin relaxation, limited averaging results in comparatively larger margins of error. Nevertheless, we are confident that the cavity VV<sup>0</sup> has a lower bound of  $T_1 \gtrsim 500 \,\mu s$ .



Figure S3 | Coherence times of cavity VV<sup>0</sup> at 6 Gauss. (a) Ramsey sequence with a  $T_2^*$  confidence interval of 95% (b) Hahn echo sequence with a  $T_2$  confidence interval of 95%



Figure S4 | Spin relaxation times of cavity VV<sup>0</sup> at 6 Gauss. (a) 100  $\mu$ s spin relaxation measurement with an exponential decay fit giving  $T_1 = 1.02 \pm 0.47$  ms. (b) 1 ms spin relaxation measurement with an exponential decay fit giving  $T_1 = 2.43 \pm 1.58$  ms. Both margins of error represent a 95% confidence interval.

### Supplementary note 8: Purcell enhancement of upper branch of cavity VV<sup>0</sup>

While the lower energy branch of the  $VV^0$  orbital fine structure displays a higher Purcell enhancement in the nanobeam cavity, it is also possible to tune the cavity into resonance with the upper branch. Doing so gives a Purcell factor of ~16, with the difference likely occurring due to the lowered spatial matching of the emission dipole with the cavity mode for the upper branch.



Figure S4 | Purcell enhancement of upper branch of VV<sup>0</sup>. (a) Emission spectrum of the VV<sup>0</sup> when excited with off-resonant 905 nm laser light with the cavity on (inset, lower right) and off (inset, upper right) resonance with the higher energy branch. A ratio of emission intensities gives a Purcell factor of ~16. The on-resonance trace for the combined plot is vertically offset for clarity.

## Supplementary note 9: Cavity VV<sup>0</sup> measurements with off-resonant optical excitation

For further characterization of the cavity  $VV^0$ , we collected both a  $g^{(2)}$  autocorrelation measurement and optically detected magnetic resonance (ODMR) using off-resonant optical excitation. The  $g^{(2)}$  measurement features antibunching behavior indicative of a single emitter, with a higher  $g^{(2)}(0)$  value of 0.374 due to increased background luminescence from other emitters, such as the NV center in SiC. The off-resonant ODMR trace features two Zeeman split resonances centered at 1.328 GHz as expected, with decreased photoluminescence (PL) when applying microwaves. This matches the behavior of a (*hh*) VV<sup>0</sup>, while the (*kk*) VV<sup>0</sup> instead displays an increase in PL under resonance<sup>1</sup>.



**Figure S5** | **Characterization of cavity VV0 with off-resonant optical excitation.** (a)  $g^{(2)}(t)$  autocorrelation measurement of the nanobeam VV<sup>0</sup> taken with off-resonant 905 nm excitation, with a best fit (red) including the presence of a nonradiative state and a horizontal line (green) at  $g^{(2)} = 0.5$  indicating the upper threshold for a single emitter. The data contains  $g^{(2)}(0) = 0.374$  with no background subtraction and the best fit line gives  $g^{(2)}(0) = 0.343$ . (b) Optically detected magnetic resonance (ODMR) collected with off-resonant 905 nm excitation under a ~25 G c-axis magnetic field to Zeeman split the resonances.

### Supplementary note 10: Discussion of cavity mode and dimensions

We used *Lumerical FDTD Solutions* in order to model the resonant modes of photonic crystal cavities, specifically for the 1D nanobeam structure. The high-Q design reported in the main text was obtained with the following dimensions:

Beam width = 415 nmBeam thickness = 322 nmHole radius = 83 nmLattice spacing (hole-to-hole) = 337 nm

The central 8 holes of the nanobeam (4 on each side of the center) are linearly tapered to ellipses with a lattice spacing and a minor axis length of 84% of the bulk values, while the major axis diameter remains unchanged. In this design the major axis is oriented perpendicular to the length along the beam. Using this design with the above dimensions and a SiC material assumed to have a constant index of refraction of 2.58, we obtained a mode with a Q of  $\sim 3 \cdot 10^5$  at a wavelength of 1131 nm, which matches the (*hh*) and (*kk*) VV<sup>0</sup> emission wavelengths. If we follow the ratio of these dimensions and scale the entire structure up or down, shorter and longer cavity resonant

wavelengths can be achieved while maintaining a Q above  $10^5$ . Thus, a structure with this approximate ratio of 1:4:4:5 for radius, lattice spacing, thickness, and width will yield a high-Q device. Outside of this particular ratio, cavity modes are supported over a wide range of dimensions and wavelengths, with quality factors typically in the  $10^4$ - $10^5$  range.

Experimentally, the radius, lattice spacing, and beam width can be freely changed in the design of the electron beam pattern while the beam thickness is fixed at a maximum of 400 nm. This is due to the thickness of the SiC grown on top of the p-doped layer, which determines the depth of the undercut. Other thicknesses could be selected for future wafer growths, but we found 400 nm to be a good balance between flexibility for thinning, structural integrity of suspended beams, and capability to support near-IR cavity resonances as-grown. In the nanobeam studied in the main text, scanning electron microscope (SEM) images of the beam gives approximate measured dimensions of:

Beam width = 450 nm Beam thickness = 400 nm Hole radius = 125 nm Lattice spacing (hole-to-hole) = 342 nm

Using these dimensions in *Lumerical* gives two modeled resonances: one at Q = 18,000 at 1077 nm, and another with Q = 22,000 at 1103 nm. SEM images also reveal a sidewall slope of roughly 86°, which has been previously modeled to blueshift cavity resonances<sup>2</sup>. Although we do not observe the modeled pair of resonances in our device, we do observe a resonance at ~1077 nm which matches the shorter wavelength. This resonance could also potentially be a blueshifted version of the 1103 nm resonance. The observed Q of 5,100 is lower than the theoretical Q's, which is common for photonic devices due to fabrication imperfections.

Additionally, this specific beam was originally designed to have a 500 nm width, a 105 nm hole radius, and a lattice spacing of 350 nm. While the lattice spacing roughly matches, discrepancies in the other dimensions can be attributed to systematic errors in fabrication and PEC etching. For example, we observe small ~5-10 nm shifts in the intended radius of holes after both the metal mask deposition/liftoff and the ICP etch of the SiC. PEC etching can also result in slight etching of the n-type or intrinsic SiC, which usually occurs after the p-type SiC has been etched away. While these systematic errors are being continuously addressed, the design of nanobeam cavities for this work was designed to have flexibility robust to this error. By fabricating a large array of cavities with varied dimensions, a subset of devices is highly likely to have a resonant mode at a desired wavelength.

#### Supplementary note 11: Photoelectrochemical etching

Photoelectrochemical (PEC) etching was used to selectively etch p-type SiC to create undercut structures. Under the proper conditions, we observe selective PEC etching to work for both NINPN and IPN doping configurations. Generally, PEC etching requires UV illumination to create electron-hole pairs in the SiC, an applied voltage across the SiC chip to bias charge carriers towards or away from the surface, and an ionic solution to both form an electrical circuit and etch away oxidized SiC at the surface. We use a 1150 mW 365 nm LED focused through a lens as a UV source and a 0.2 M KOH mixture as an ionic solution. We apply voltage across the SiC chip between an Ohmic contact on the backside n-type surface and a platinum wire submerged in the KOH solution. A schematic of the PEC etching setup can be seen in Fig. s6.



Figure S6 | Photoelectrochemical etching setup. A 4H-SiC sample (here with NINPN doped layers) is submerged in a 0.2 M KOH solution. electrical contacts are made with an electrical wire to Ohmic NiCr on the back of the chip and a platinum wire in solution. The voltage is varied so as to give a 5-10  $\mu$ A photocurrent and is typically within a ±1 V range, although it could be positive or negative. A 365 nm UV LED is used as a UV source.

The voltage is set in order to achieve 5-10  $\mu$ A of photocurrent, which is defined as the change in current that is observed when the UV source is turned on. Depending on the sample, this voltage lies within a ±1 V range. We find the 5-10  $\mu$ A of photocurrent to be an ideal balance to achieve etch selectivity while maintaining a reasonable etch rate. PEC etch rates are typically on the order of 100 nm/hour, so a total etch time of 5-6 hours is typical for a sample.

## Supplementary note 12: g<sup>(2)</sup> rate equations

We model the  $VV^{0}$ 's photoluminescence according to simplified three level system, as given in Fig. S5 below.



Figure S7 | Three-level model of VV<sup>0</sup> optical transitions. In this diagram,  $n_0$  represents the ground state,  $n_1$  represents the excited state, and  $n_2$  represents an overall nonradiative state, which in practice is a combination of the inter-system crossing singlet state and ionized states of the VV<sup>0</sup>. Rates between these levels are denoted by  $g_{01}$ ,  $g_{10}$ ,  $g_{12}$ , and  $g_{20}$ .

Here,  $n_0$  represents the VV<sup>0</sup> ground state,  $n_1$  represents the VV<sup>0</sup> excited state, and  $n_2$  represents all intermediate states involved in nonradiative decays (e.g., different charge states and the intersystem crossing). The  $g_{01}$  rate is the optical pumping rate, and depends on the laser power. The  $g_{10}$  rate is the radiative decay rate, measured to be  $15.7 \pm 0.3$  ns in the main text without Purcell enhancement and  $5.3 \pm 0.1$  ns with enhancement. The  $g_{12}$  rate is the same as the  $\tau_{dark}$  state referred to in the main text, which represents all nonradiative transitions out of the excited state. The  $g_{20}$  rate represents all transitions from an intermediate state (such as a charged VV state or the VV<sup>0</sup> ISC state) back to the VV<sup>0</sup> ground state. We do not measure the  $g_{20}$  rate directly, although it plays a role in the overall photoluminescence brightness of the VV<sup>0</sup>.

According to this three-level model, the dynamics of this system can be expressed as a system of differential equations:

$$n'_{0}(t) = -g_{01}n_{0}(t) + g_{10}n_{1}(t) + g_{20}n_{2}(t)$$
$$n'_{1}(t) = g_{01}n_{0}(t) - g_{10}n_{1}(t) - g_{12}n_{1}(t)$$
$$n'_{2}(t) = -g_{20}n_{2}(t) + g_{12}n_{1}(t)$$

By setting the initial conditions set at the ground state:

$$n_0(0) = 1$$
,  $n_1(0) = 0$ ,  $n_2(0) = 0$ 

We obtain a solution to the excited state population  $n_1(t)$  using Wolfram Mathematica, which gives:

$$n_1(t) = \frac{g_{01} \exp\left(-\frac{1}{2}(a_1+b)t\right) \left(a_3(-1+\exp(bt)) + g_{20}b\left(-1-\exp(bt) + 2\exp\left(\frac{1}{2}(a_1+b)t\right)\right)\right)}{2a_2b}$$

Where:

$$a_{1} = g_{01} + g_{10} + g_{12} + g_{20}$$

$$a_{2} = g_{01}g_{12} + g_{20}(g_{01} + g_{10} + g_{12})$$

$$a_{3} = g_{20}(g_{10} + g_{12} - g_{20}) + g_{01}(2g_{12} + g_{20})$$

$$b = \sqrt{a_{1}^{2} - 4a_{2}}$$

Plotting this function, where |t| is used as the input, gives the  $g^{(2)}$  fits used in the main text and here in the supplement. For the  $g^2$  plot in the main text, the fit returns a  $g_{12}$  value of 16.48 ps<sup>-1</sup>, which corresponds to a lifetime of  $\tau_{dark} = 1/g_{12} = 60.7 ns$ .

### Supplementary note 13: Expressions for the Purcell factor

### 13.1 Purcell factor in terms of cavity-emitter properties

The full equation for the Purcell factor, including cavity-emitter matching, is given by:

$$F = \left(\frac{\left|\vec{\mu} \cdot \vec{E}\right|}{\left|\vec{\mu}\right| \left|\vec{E}_{max}\right|}\right)^{2} \left(\frac{1}{1 + 4Q^{2} \left(\frac{\lambda_{ZPL}}{\lambda_{cavity}}\right)^{2}}\right) \frac{3Q}{4\pi^{2}V} \left(\frac{\lambda_{cavity}}{n}\right)^{3} + 1$$

Where  $\vec{\mu}$  is the electric dipole moment of the emitter,  $\vec{E}$  is the electric field from the emitter,  $\vec{E}_{max}$  is the maximum electric field from the cavity mode,  $\lambda_{ZPL}$  is the wavelength of the ZPL,  $\lambda_{cavity}$  is the resonant wavelength of the cavity, Q is the cavity quality factor, V is the cavity mode volume, and n is the index of refraction of the material. The first term  $\left(\frac{|\vec{\mu} \cdot \vec{E}|}{|\vec{\mu}||\vec{E}_{max}|}\right)^2$  represents spatial overlap between the emitter and cavity mode, where both the position and orientation of the emitter play important roles for the overall coupling. The second term  $1/\left(1 + 4Q^2 \left(\frac{\lambda_{ZPL}}{\lambda_{cavity}}\right)^2\right)$  represents spectral matching between the emitter and cavity. For higher Q cavities it becomes increasingly critical for the emitter wavelength to be matched with the cavity resonance. In the case of ideal

critical for the emitter wavelength to be matched with the cavity resonance. In the case of ideal coupling, both of these factors are one, and so the approximation  $F \approx Q/V$  is commonly used to highlight the importance of high quality factors and small mode volumes.

### 13.2 Purcell factor in context of this work

For the purposes of this work, the definition of a Purcell factor as a ratio of emission rates<sup>3</sup> applies to the zero-phonon line transition:

$$F = \frac{\Gamma_{ZPL,on}}{\Gamma_{ZPL,off}} = \frac{\tau_{ZPL,off}}{\tau_{ZPL,on}}, \qquad (1)$$

Where  $\Gamma_{ZPL,on}$  and  $\Gamma_{ZPL,off}$  are the ZPL emission rates on and off cavity resonance, while  $\tau_{ZPL,on}$  and  $\tau_{ZPL,off}$  are the ZPL lifetimes on and off cavity resonance. It's worth noting that equation (1) is equivalent to the statement that on resonance, the lifetime  $\tau_{ZPL}$  is modified as:

$$\tau_{ZPL,on} = \frac{\tau_{ZPL,off}}{F},\qquad(2)$$

With this in mind, we can write expressions for the total optical lifetime of the  $VV^0$  emitter when on and off cavity resonance. When off resonance, the lifetime will be given by a combination of the ZPL transition, all phonon sideband (PSB) transitions, and all non-radiative dark state transitions:

$$\frac{1}{\tau_{off}} = \frac{1}{\tau_{ZPL}} + \frac{1}{\tau_{PSB}} + \frac{1}{\tau_{dark}},\qquad(3)$$

Where  $\tau_{ZPL}$  is the lifetime of the ZPL off cavity resonance,  $\tau_{PSB}$  is the combined lifetime of all transitions into the phonon sideband,  $\tau_{dark}$  is the combined lifetime of all nonradiative decays, and  $\tau_{off}$  is the measured lifetime of the VV<sup>0</sup> when off resonance with the cavity. When the cavity is tuned into the resonance with the VV<sup>0</sup> ZPL, the overall lifetime will be modified as given in eq. (2):

$$\frac{1}{\tau_{on}} = \frac{F}{\tau_{ZPL}} + \frac{1}{\tau_{PSB}} + \frac{1}{\tau_{dark}},\qquad(4)$$

Where  $\tau_{on}$  is the measured lifetime of the VV<sup>0</sup> when on resonance with the cavity. Lastly, for upcoming derivations we will need an expression for the Debye-Waller (DW) factor. Given that this factor is the ratio of emitted light into the ZPL, we can express it in terms of lifetimes as:

$$\alpha = \frac{\tau_{PSB}}{\tau_{ZPL} + \tau_{PSB}}, \qquad \text{Debye Waller factor,} \qquad (5)$$

Equations 1-5 can then be used to derive the Purcell factor equations listed in the main text, as will be outlined below.

### 13.3 Purcell factor in terms of lifetimes

Eq. (3) of the main text expresses the Purcell factor in terms of measurable lifetimes. To obtain this equation, we start by rearranging Eq. (2) to isolate the Purcell factor F:

$$F = \tau_{ZPL} \left( \frac{1}{\tau_{on}} - \frac{1}{\tau_{PSB}} - \frac{1}{\tau_{dark}} \right), \qquad (6)$$

While this equation is analytically correct, it is difficult to directly measure values for  $\tau_{ZPL}$  and  $\tau_{PSB}$ . To obtain these terms we can rearrange Eqs. (3) and (5) to give:

$$\tau_{ZPL} = \frac{\tau_{off} \tau_{dark} \tau_{PSB}}{(\tau_{dark} - \tau_{off}) \tau_{PSB} - \tau_{off} \tau_{dark}}, \quad (7)$$
$$\tau_{PSB} = \frac{\tau_{ZPL} \alpha}{1 - \alpha}, \quad (8)$$

Then, after substituting (7) into (8), we can rewrite  $\tau_{ZPL}$  and  $\tau_{PSB}$  in terms of experimentally measurable quantities:

$$\tau_{ZPL} = \frac{\tau_{dark} \tau_{off}}{\alpha (\tau_{dark} - \tau_{off})}, \qquad (9)$$

$$\tau_{PSB} = \frac{\tau_{dark} \tau_{off}}{(1 - \alpha) (\tau_{dark} - \tau_{off})}, \qquad (10)$$

Substituting (9) and (10) into (6) then gives F in terms of experimentally measurable quantities:

$$F = \frac{\tau_{dark} (\tau_{off} - \tau_{on})}{\alpha \tau_{on} (\tau_{dark} - \tau_{off})} + 1, \qquad (11)$$

This is eq. (3) in the main text. It is also consistent with no enhancement corresponding to F = 1, as the quantity  $(\tau_{off} - \tau_{on})$  goes to zero.

### 13.4 Purcell factor in terms of Debye-Waller Factor

Eq. (4) of the main text expresses the Purcell factor in terms of off- and on-resonance DW factors. To obtain this expression, we start with eq. (5) of this supplement:

$$\alpha = \frac{\tau_{PSB}}{\tau_{ZPL} + \tau_{PSB}}, \qquad \text{Debye Waller factor,} \qquad (5)$$

And note that on resonance, we have:

$$\tau_{ZPL} \to \frac{\tau_{ZPL}}{F}, \qquad \beta = \frac{\tau_{PSB}}{\frac{\tau_{ZPL}}{F} + \tau_{PSB}}$$

Where  $\beta$  is the Debye-Waller factor on resonance. Substituting eq. (8) into this expression and rewriting gives:

$$\beta = \frac{F\alpha}{1 - \alpha + F\alpha},\qquad(12)$$

Isolating F then gives:

$$F = \frac{\beta(\alpha - 1)}{\alpha(\beta - 1)},$$
 (13)

Which matches eq. (4) from the main text. Incidentally, eq. (12) can be used to estimate an enhanced DW factor for a given a Purcell factor, which is also done in the main text.

A secondary estimate of the Purcell factor and enhanced DW factor is also done using the total off-resonant excitation (905 nm) counts on and off cavity resonance. Based on observed background subtracted count rates of 120 and 460 kCts/s, we say:

$$\alpha = \frac{ZPL_{off}}{total_{off}} = 0.053, \quad total_{off} = 120$$

$$\beta = \frac{ZPL_{on}}{total_{on}}, \quad total_{on} = 460$$

Where the numbers of 120 and 460 are meant to serve as relative measures for PL for off and on cavity resonance, and are chosen to be unitless for clarity. Continuing with this convention, the equation for  $\alpha$  can be rearranged to give:

$$ZPL_{off} = \alpha \cdot total_{off} = 0.053 \cdot 120 = 6.36$$

If we assume the increase of total counts on cavity resonance is solely due to increased counts into the ZPL, then we can say:

$$ZPL_{on} = ZPL_{off} + (total_{on} - total_{off}) = 6.36 + 340 = 346.36$$

In total, the enhanced DW factor then matches the number given in the main text:

$$\beta = \frac{ZPL_{on}}{total_{on}} = \frac{346.36k}{460k} = 75.3\% \approx 75\%$$

Lastly, in the main text this number is used to obtain another measure of the Purcell factor. In this case, we reformulate eq. (1):

$$F = \frac{\Gamma_{ZPL,on}}{\Gamma_{ZPL,off}} = \frac{ZPL_{on}}{ZPL_{off}}$$

Which, using the numbers already obtained, gives:

$$F = \frac{346.36k}{6.36k} = 54.46 \approx 54$$

Which gives the number reported in the main text.

## Supplementary note 14: Effect of strain on VV<sup>0</sup>

### 14.1 Attribution of strain to nanobeam VV<sup>0</sup>

We attribute differences in both ZPL and ODMR between the nanobeam divacancy in the main text and a bulk (*hh*)  $VV^0$  to the existence of high strain around the defect, as strain can result in shifts in both the ground state and excited state Hamiltonians. Electric fields may also play a role given the proximity of n-doped layers, but the field strength should not be significant due to the symmetric configuration of doping. Additionally, previous work has only observed ~1 nm ZPL shifts and no ODMR change under the presence of built-in electric fields from a PIN SiC diode<sup>4</sup>. We also observe similar ZPL and ODMR shifts for defects in unpatterned regions of the NIN epilayer (see supplementary note 6), indicating that the attributed strain is present throughout the sample. This suggests that existing strain is due to growth conditions rather than fabricated nanostructures. More specifically, the incorporation of highly doped layers during growth of the NINPN epilayers is known to generate strain in the SiC lattice<sup>5–8</sup>. Given that intrinsically doped SiC can be grown while still maintaining a selectivity for a PEC undercut, it should be possible to lower the n-dopant levels to mitigate these effects in future work.

To quantify the amount of strain in the VV<sup>0</sup>'s local environment, we incorporate the observed shift of the ODMR frequency into an analysis of the ground-state spin Hamiltonian (see below, 14.2). This produces calculated strain values of  $\varepsilon_{xx} = \varepsilon_{yy} = -4.61 \cdot 10^{-4}$  and  $\varepsilon_{zz} = 3.64 \cdot 10^{-4}$  with zero shear, which is consistent with strain magnitudes found in SiC nanoparticles<sup>9</sup>. Given the similarity of spin-strain coupling dynamics between this system and the NV center in diamond, we can also obtain an approximate effect of this strain on the excited state using the NV center parameters as a proxy<sup>10</sup> (see below, 14.3). This results in a predicted ZPL shift of -2.7 THz, which is within a factor of five of the -13 THz ZPL shift for the VV<sup>0</sup> observed here. This factor is likely the result of comparatively higher excited state strain coupling parameters for the VV<sup>0</sup>. For example, in previous spin-strain work Lee *et al.* reported a ~850 THz/strain splitting for the  $|E_x\rangle$  and  $|E_y\rangle$  states in the NV center<sup>10</sup>, while Falk *et al.* reported a larger ~2,900 THz/strain splitting for the observed defect behavior to a (hh) VV<sup>0</sup> with strain on the order of 10<sup>-4</sup>.

## 14.2 Effect of strain on VV<sup>0</sup> Hamiltonian and calculated strain values

To examine the effect of strain on  $VV^0$  behavior, it is useful to examine the spin basis for the  $VV^0$  ground state. The ground state spin-spin Hamiltonian for trigonal defects, ignoring the effects of nuclear spins and applied fields, is given by<sup>12</sup>:

$$\widehat{H}_{gs} = \begin{bmatrix} D & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D \end{bmatrix}$$

Where

$$D = \frac{3}{2}D_{zz}, \qquad E = \frac{1}{2}(D_{xx} - D_{yy})$$

Are determined from diagonal elements of the zero-field splitting tensor  $\vec{D}$ . Typically for c-axis (*hh*) and (*kk*) defects, no splitting of the ODMR spectrum is observed at zero-field, meaning the *E* term is zero. In the basal (*hk*) and (*kh*) defects, however, a significant zero-field ODMR splitting is observed. This corresponds *E* values of 82.0 MHz and 18.7 MHz for the (*hk*) and (*kh*) VV<sup>0</sup>s, respectively<sup>13</sup>. For the cavity VV<sup>0</sup> studied in this work, we observe no transverse zero-field splitting, prompting us to use set the *E* term to zero:

$$\widehat{H}_{gs} = \begin{bmatrix} D & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D \end{bmatrix}$$

This Hamiltonian gives eigenvalues and eigenvectors of:

$$\lambda_{1} = 0, \qquad v_{1} = \begin{pmatrix} 0\\1\\0 \end{pmatrix}, \qquad m_{s} = |0\rangle$$
$$\lambda_{2} = D, \qquad v_{2} = \begin{pmatrix} 0\\0\\1 \end{pmatrix}, \qquad m_{s} = |-1\rangle$$
$$\lambda_{3} = D, \qquad v_{3} = \begin{pmatrix} 1\\0\\0 \end{pmatrix}, \qquad m_{s} = |+1\rangle$$

Here the  $|-1\rangle$  and  $|+1\rangle$  states are degenerate under zero magnetic field, which manifests itself as a single resonance under an ODMR scan. As we add strain to this system, *D* becomes modified due to changes in the  $\vec{D}$  tensor. To quantify the full effect of strain on the elements of  $\vec{D}$ , it is necessary to use the spin-strain coupling tensor  $\vec{G}$ , which follows the relation<sup>14</sup>:

$$\begin{pmatrix} \Delta D_{xx} \\ \Delta D_{yy} \\ \Delta D_{zz} \\ \Delta D_{yz} \\ \Delta D_{xz} \\ \Delta D_{xy} \end{pmatrix} = \vec{G} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ 2\varepsilon_{yz} \\ 2\varepsilon_{yz} \\ 2\varepsilon_{xz} \\ 2\varepsilon_{xy} \end{pmatrix}, \quad (14)$$

Or, more explicitly,

$$\begin{pmatrix} \Delta D_{xx} \\ \Delta D_{yy} \\ \Delta D_{yz} \\ \Delta D_{yz} \\ \Delta D_{yz} \\ \Delta D_{xz} \\ \Delta D_{xy} \end{pmatrix} = \begin{pmatrix} G_{11} & G_{12} & G_{13} & G_{14} & 0 & 0 \\ G_{12} & G_{11} & G_{13} & -G_{14} & 0 & 0 \\ -G_{11} - G_{12} & -G_{11} - G_{12} & -2G_{13} & 0 & 0 & 0 \\ G_{41} & -G_{41} & 0 & G_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & G_{44} & G_{41} \\ 0 & 0 & 0 & 0 & 0 & G_{14} & \frac{G_{11} - G_{12}}{2} \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ 2\varepsilon_{yz} \\ 2\varepsilon_{xz} \\ 2\varepsilon_{xy} \end{pmatrix}$$

Where  $\Delta D_{ij}$  represent changes in elements of  $\vec{D}$  due to strain terms  $\varepsilon_{ij}$ . The individual terms of  $\vec{G}$  have not been experimentally measured for c-axis divacancies in silicon carbide, but have been computed using DFT calculations to match observed D values<sup>14</sup>. The  $D = \frac{3}{2}D_{zz}$  term determines the center of the zero-field ODMR spectrum, which is 1.328 GHz for our sample. The shift from the expected 1.336 GHz value for a (hh) VV<sup>0</sup> then gives  $\Delta D_{zz} = -5.33$  MHz. To fully solve for the expected strain, we also set  $\Delta E = \frac{1}{2} (\Delta D_{xx} - \Delta D_{yy})$  to zero to be consistent with our observations. We can also impose the fact that  $\vec{D}$  is traceless to say  $D_{xx} + D_{yy} + D_{zz} = 0$ . Lastly, we set the mixed  $\vec{D}$  terms to zero, meaning  $\Delta D_{yz} = \Delta D_{xz} = \Delta D_{xy} = 0$ , as we do not expect high levels of shear in our sample. The combination of these constraints then gives a unique solution for the strain. Using Eq. (14), we obtain the strain values of:

$$\varepsilon_{xx} = \varepsilon_{yy} = -4.61 \cdot 10^{-4}, \qquad \varepsilon_{zz} = 3.64 \cdot 10^{-4}, \qquad \varepsilon_{yz} = \varepsilon_{xz} = \varepsilon_{xy} = 0$$

It is worth noting that despite these strain values, the  $m_s = -1,0,+1$  basis remained unchanged due to the unchanging  $E = \frac{1}{2} (D_{xx} - D_{yy}) = 0$  term.

### 14.3 Corroboration of strain with diamond NV center parameters

To corroborate the plausibility of these strain magnitudes, we can examine their approximate effect on the excited state. For defects with  $C_{3v}$  symmetry, strain is known to both shift and split the frequency of the zero-phonon line<sup>15</sup>. While the excited state strain coupling constants for the  $VV^0$  are not known, they have been measured for the NV center in diamond<sup>10</sup> and we expect the  $VV^0$  excited state spin strain coupling to follow similar dynamics. Using the  $\{|A\rangle, |E_x\rangle, |E_y\rangle$  states as a basis, the excited state NV center Hamiltonian can be written as<sup>10,15</sup>:

$$H_{strain} = [f_{ZPL} + \lambda_{A_1}\epsilon_{zz} + \lambda_{A'_1}(\epsilon_{xx} + \epsilon_{yy})] \times [|E_x\rangle\langle E_x| + |E_y\rangle\langle E_y|]$$
$$+ [\lambda_E(\epsilon_{yy} - \epsilon_{xx}) + \lambda_{E'}(\epsilon_{xz} + \epsilon_{zx})] \times [|E_x\rangle\langle E_x| - |E_y\rangle\langle E_y|]$$
$$+ [\lambda_E(\epsilon_{xy} + \epsilon_{yx}) + \lambda_{E'}(\epsilon_{yz} + \epsilon_{zy})] \times [|E_x\rangle\langle E_y| + |E_y\rangle\langle E_x|]$$

Where the orbital-strain-coupling constants  $\lambda_{A_1}$ ,  $\lambda_{A'_1}$ ,  $\lambda_E$ , and  $\lambda_{E'}$  have been measured for the NV center to be:

$$\begin{split} \lambda_{A_1} &= -1.95 \pm 0.29 \; PHz/strain \\ \lambda_{A_1'} &= 2.16 \pm 0.32 \; PHz/strain \\ \lambda_E &= -0.85 \pm 0.13 \; PHz/strain \\ \lambda_{E'} &= 0.002 \pm 0.01 \; PHz/strain \end{split}$$

For our calculated strain values, only the shift in ZPL will be nonzero, corresponding to the first term:

$$H_{strain} = \left[ f_{ZPL} + \lambda_{A_1} \epsilon_{zz} + \lambda_{A'_1} (\epsilon_{xx} + \epsilon_{yy}) \right] \times \left[ |E_x\rangle \langle E_x| + |E_y\rangle \langle E_y| \right]$$

Plugging in our predicted strain values then gives:

$$\Delta f_{ZPL} = \lambda_{A_1} \epsilon_{zz} + \lambda_{A'_1} (\epsilon_{xx} + \epsilon_{yy}) = -1950 \cdot \epsilon_{zz} + 2160 (\epsilon_{xx} + \epsilon_{yy}) THz$$
$$\Delta f_{ZPL} = (-1950(3.639 \cdot 10^{-4}) + 2160(2 \cdot -4.611 \cdot 10^{-4})) THz$$
$$\Delta f_{ZPL} \approx -2.7 THz$$

Which is the value reported above in 14.1.

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