Selective Purcell enhancement of two closely linked zero-phonon transitions of a silicon carbide color center

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Point defects in silicon carbide are rapidly becoming a platform of great interest for single-photon generation, quantum sensing, and quantum information science. Photonic crystal cavities (PCCs) can serve as an efficient light–matter interface both to augment the defect emission and to aid in studying the defects’ properties. In this work, we fabricate 1D nanobeam PCCs in 4H-silicon carbide with embedded silicon vacancy centers. These cavities are used to achieve Purcell enhancement of two closely spaced defect zero-phonon lines (ZPL). Enhancements of \(\sim 80\)-fold are measured using multiple techniques. Additionally, the nature of the cavity coupling to the different ZPLs is examined.

results

The factor \(\xi\) takes into account the spectral and spatial overlap of the cavity field with the emitter being enhanced, as well as the alignment of the cavity field and the emitter’s optical dipole. With perfect overlap and alignment, \(\xi = 1\). Thus, for a given \(\xi\), maximizing emission enhancement depends upon fabricating cavities with high \(Q\) and low \(V\). To provide high \(Q/V\), the PCC design we have chosen consists of a linear array of holes (Fig. 1A), where the eight innermost holes on either side of the center are linearly tapered to define a cavity region. The holes are tapered in both their center-to-center spacing and their parallel-to-beam diameter (\(x\) axis in Fig. 1B), creating elliptical holes in this

Significance

Semiconductor point defects have shown great promise in their application to quantum information and sensing in the solid state. However, it is an ongoing challenge to efficiently access the light emitted by these spin-active defects and, furthermore, to enhance the emission at wavelengths that can be used to create indistinguishable photons. Such emission enhancement can be achieved by placing the defects within optical microcavities. Here, using 1D photonic crystal cavities, we report the significant enhancement of point-defect emission in silicon carbide, which hosts a suite of intriguing spin-active defects. In addition to measuring large enhancements, we also demonstrate how the cavity coupling can potentially allow access to a variety of information about the defects and their environment.
inner region. For both parameters, the minimum tapered value is 84% of the values in the outer holes. Finite-difference time-domain (FDTD) simulations (Lumerical Solutions) confirm that these cavities have high theoretical $Q$ of $4 \times 10^6$ and small mode volume of 0.45 $(\lambda/n)$ (3), where $\lambda$ is the resonant wavelength of the cavity mode and $n$ the refractive index. The simulated electric-field profile of the primary transverse electric (TE) mode is shown Fig. 1B, showing that the fields are well localized to the inner, tapered cavity region.

The fabrication technique we used, described in Materials and Methods, resulted in chips with ~3,000 cavities, designed with cavity resonances that span the full range of the SiV emission. The point defects were introduced after the fabrication of the cavities: The wafers with PCCs were ion implanted (Cutting Edge Ions Inc; Materials and Methods) without postannealing to create ensembles of SiV centers. Based on comparison with other work (9, 28) quantifying the creation of these centers in 4H-SiC, we expect a few tens (~10–30) of defect centers within the cavity mode volume. Photoluminescence (PL) spectra of the implanted samples taken at 4 K in a helium-cooled continuous-flow cryostat (Janis) show the positions of the ZPLs of both the V1 and V2 types of the SiV center. These two centers correspond to the two inequivalent Si lattice sites in 4H-SiC, with V1 belonging to the cubic (k) site and V2 to the hexagonal (h) site (12). The defects at both sites have total spin $S = 3/2$ and show optically detected magnetic resonance signals (3, 4, 12). As seen in Fig. 2, the V1 ZPL is observed at 861.4 nm and the V2 ZPL at 916.5 nm, corresponding to values previously reported in literature. Moreover, the V1 center is known to have a high-temperature companion ZPL, typically denoted V1', stemming from a transition to an excited state of slightly higher energy than the excited state corresponding to the V1 transition (13). Because of the 4.4-meV energy splitting between the V1' and V1 excited states (schematic shown in Fig. 2E), the intensities of the corresponding ZPLs are temperature dependent (13). This is clearly observed in the spectra shown in Fig. 2: When the excitation laser is at a power of 100 µW, emission into the V1' line is suppressed (Fig. 2C and D). However, at a higher excitation laser power of 600 µW, the V1' transition intensity is nearly comparable to that of the V1 peak due to laser-induced heating (Fig. 2A and B). At the higher laser power, the branching ratio into each ZPL is ~1–1.3%, whereas at lower laser power the branching ratio into the V1 line is ~1.6% with only ~0.5% into the V1' line.

After ion implantation, we characterized the fabricated PCCs using confocal PL microscopy. A pulsed Ti:Sapphire laser tuned to a wavelength of 760 nm was used to optically excite the SiV centers in the cavities, and the emitted light was passed to a spectrometer for analysis. Measured PCC spectra showed cavity modes decorating the defects' broad PL signal (consisting primarily of phonon sideband emission). Even without postimplantation annealing of the material, the cavities displayed high values of $Q$, typically between ~2,000 and 5,000 (Fig. 1C), as calculated by fitting the cavity spectrum to a Lorentzian profile. This disparity
which have the same ground state and two different excited states. The results of this tuning with one such cavity using an excitation power of 600 μW are shown in Fig. 3. An intensity map as a function of tuning step in the vicinity of the V1 and V1’ ZPLs is shown in Fig. 3A. The excitation power and collection time were kept constant for each spectrum acquired so that the intensities could be directly compared. As labeled, the two fixed lines correspond to the V1 and V1’ ZPLs, through which the cavity mode sweeps as it is redshifted by the condensed gas. The intensity of each ZPL reaches a maximum when it coincides with the cavity mode wavelength.

Additionally, Fig. 3B–D provides details of the emission spectra at specific tuning steps to better underscore the increased emission into the ZPLs when they are resonant with the cavity mode. In Fig. 3B, we see the cavity mode far off-resonance from both ZPLs. At cryogenic temperatures, the defects emit very little light at the cavity wavelength, and therefore emission into the ZPLs is of low intensity. Fitting the spectrum reveals a cavity Q of ~2,200. The corresponding FWHM of the cavity, 0.4 nm, is well matched to the ZPL linewidths (on the order of ~0.5 nm), which aids in efficient coupling. In Fig. 3C and D, the cavity mode is fully on-resonance with the V1’ and V1 ZPLs, respectively. The large corresponding increases in intensity are readily apparent and we moreover see that the on-resonance intensity of the V1’ is significantly higher than that of the V1. A typical means of quantifying the degree of Purcell enhancement, P, achieved by a

between measured and theoretical Q is commonly observed in fabricated PCC devices (16–18, 20, 23). As discussed in previous work (25), the measured Q is often limited by fabrication imperfections, particularly those linked to reactive ion etching, like roughness, angled sidewalls, and other ion-induced damage. Other limiting factors may include absorption by the SiVs introduced to the devices and by other defects also created by the ion implantation. Using Q of 2,000 and the simulated V of ~0.45 (ℏ/π) (3) in Eq. 1, we calculate a maximum Purcell enhancement of ~340, assuming ξ of 1. Even without perfect defect placement within the cavity, significant ZPL enhancement can be achieved, as long as the overlap of the defect spatial location and cavity field maximum is sufficient to yield ξ of ~0.1 or greater.

To demonstrate such enhancement, it is easiest to choose devices with modes that are slightly blueshifted from the desired ZPL wavelength and then slowly redshift the cavity mode through resonance, using nitrogen gas condensation (18). This technique is commonly used and can typically reversibly redshift cavity modes by at least 20 nm without degrading their Qs (17, 18, 22, 29). Modes can also be appropriately blueshifted of the ZPL by using previously reported techniques for tuning 4H-SiC cavities (25). Therefore, we selected cavities with well-coupled modes that were blueshifted relative to the 859.1-nm V1’ line and took measurements as these modes are tuned through both V1’ and V1 ZPLs. By focusing on the interaction with these two ZPLs, we use the cavity as an exquisitely sensitive probe of these transitions
cavity is to compare the on- and off-resonant ZPL intensity (17, 18). Indeed, with the cavity off-resonance, the total defect emission rate is given by (17)

\[
\frac{1}{\tau_{\text{off}}} = \frac{1}{\tau_{\text{ZPL}}} + \frac{1}{\tau_{\text{PSB}}}
\]

where \(1/\tau_{\text{ZPL}}\) is the emission rate into the zero-phonon line and \(1/\tau_{\text{PSB}}\) is the emission rate into the phonon sideband. This off-resonance emission rate within the cavity is still significant, but it may be reduced compared with the defect emission rate in bulk (as demonstrated later) due to the photonic bandgap effect (16). On-resonance, the cavity enhances the emission rate into the ZPL such that the total defect emission rate is given by

\[
\frac{1}{\tau_{\text{on}}} = \frac{F + \Gamma}{\tau_{\text{ZPL}}} + \frac{1}{\tau_{\text{PSB}}}
\]

Therefore, as previously argued (17), the on- and off-resonant ZPL intensity can be compared with estimate of the Purcell enhancement of the defects by the relation

\[
F = \frac{I_{\text{ZPL,off}}}{I_{\text{ZPL,off}}} - 1.
\]

From this equation, we obtain an enhancement of 75 for the V1' ZPL and 22 for the V1 ZPL in the cavity described in Fig. 3. We then carried out the same cavity tuning but using a lower excitation power of 100 µW. These data are shown in Fig. 4, with Fig. 4 B–D showing the detailed spectra of the cavity mode far off-resonance and then on-resonance with the V1 and V1' ZPLs. Using on- and off-resonance intensity measurements, we derive the same degree of enhancement as observed for the higher-power (600-µW) excitation: an enhancement of 75 for the V1' ZPL and 22 for the V1 ZPL. However, although the cavity enhancement for both transitions is thus shown to be independent of excitation power used, the ratio of the on-resonance intensities for the V1' vs. V1 ZPLs is much larger in the 600-µW (Fig. 3 C and D) case than in the 100-µW case (Fig. 4 C and D). This arises from the fact that the relative intensity ratio of the V1'/V1 ZPLs at 600 µW (~1.3x), compared with at 100-µW excitation (~3x), is the same whether on- or off-resonance. Thus, the cavity is quite specific in its enhancement of these two closely spaced and related transitions. Furthermore, by controlling both the excitation power and the ZPL to which the cavity is coupled, maximum contrast of emission into one ZPL or the other as desired can be achieved.

To confirm that we are achieving the spontaneous emission enhancement that is characteristic of the Purcell factor, we present further measurements on a second cavity, which showed the largest measured Purcell enhancement. We begin by considering the spontaneous emission factor, \(\beta\), which can be calculated as the fraction of light emitted into the ZPL with the cavity mode on-resonance, \(I_{\text{ZPL,on}}/I_{\text{total}}\) (28). We can relate \(\beta\) to the Purcell factor by the equation (30)

\[
\beta = \frac{\Gamma}{1 + (F - 1)},
\]

where \(\Gamma\) is the branching ratio into the ZPL when the cavity is off-resonance. This can be rearranged to give

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

Fig. 5A shows the full PL spectra of this cavity. The upper and lower spectra show the cavity on-resonance with the V1' and V1 ZPLs, respectively. Fig. 5A (Inset) shows the off-resonance spectrum. In this cavity, for the V1' (V1) transitions, the values of \(\beta\) are 47.4% (23.7%), and the values of \(\Gamma\) are 1% (1.2%). These correspond to \(F \sim 89\) and \(\sim 25\) for the V1' and V1 ZPLs, respectively, which match the values derived by comparing the total intensity of the ZPLs on and off-resonance, \(\sim 90\) and \(\sim 26\). Furthermore, these measurements demonstrate that the ZPL is being selectively enhanced without a corresponding increase in the sideband emission, as desired. Therefore, using these cavities, we are able to direct nearly half of the SiV emission into the V1' ZPL, as opposed to the natural branching ratio of \(\sim 1\%\). Achieving such an increase is extremely important for quantum information applications using these point defects (e.g., entanglement) that can only make use of ZPL emission (15–17).
We further corroborate the spontaneous emission enhancement by measuring the excited-state lifetime of the SiV defect on-resonance with the cavity. The Purcell enhancement can be calculated from lifetime measurements using the following relation (16, 29, 30):

\[ F = \frac{\tau_0}{\tau} = \frac{1}{1 - \frac{\tau_0}{\tau_{\text{eff}}} - \frac{1}{\tau_{\text{off}}}}. \]

Here, \( \tau_{\text{on}} \) refers to the lifetime when the mode is on-resonance with the ZPL, \( \tau_{\text{off}} \) the lifetime when the mode is off-resonance, and \( \tau_0 \) the natural lifetime in the bulk material. When on-resonance, the fluorescence lifetime decreases from \( \tau_0 \) due to the increased photonic density of states. When the cavity analyzed in Fig. 5A was tuned into resonance with the V1’ ZPL, the ZPL emission was selected using a narrow bandpass filter (860-nm center wavelength, 10-nm FWHM) and passed to an avalanche photodiode for single-photon counting. Fig. 5B shows the resultant fluorescence decay signal (green curve), which is fit to yield an on-resonance lifetime of 4.2 ns. We also measure an off-resonance lifetime of 9.4 ns (blue curve) and a bulk lifetime of 6.4 ns (red curve). The bulk lifetime matches previous measurements of the silicon vacancy fluorescence lifetime (6). Moreover, as mentioned previously, the off-resonance lifetime is longer than the bulk lifetime due to a reduced density of states when the cavity mode is off-resonance with the ZPL. Using these values yields an estimated Purcell enhancement of 84, which substantiates the value estimated from measurements of ZPL intensity enhancement and increased spontaneous emission fraction into the ZPL.

Discussion and Conclusion

Lastly, we further explore the different enhancement factors for the V1’ ZPL compared with the V1 ZPL. The ~3.5-fold greater enhancement of the V1’ ZPL in both cavities presented in this work is also recapitulated in measurements of all other cavities tuned through resonance. Because we are probing ZPLs that are very close spectrally, the \( Q \) and \( V \) of a given cavity change negligibly as it is tuned through resonance with both ZPLs. Moreover, because the ZPLs result from transitions of the same SiV centers (those sitting at the \( k \) lattice sites within the cavity volume), the emitter-field spatial overlap of the ensemble of defects being enhanced in a given cavity does not vary for the two ZPLs. Therefore, the most likely explanation for the difference in enhancement between the V1’ and V1 ZPLs is a difference in alignment between the cavity field vector and the emitter optical dipole. Our SiC crystal is grown along the c axis, and therefore the plane of the PCCs (and correspondingly the cavity mode polarization) is perpendicular to the c axis. Previous work has suggested that the V1’ transition is strongly polarized perpendicular to the c axis and the V1 transition is weakly polarized parallel to the c axis (13). Such polarization would explain the difference in enhancement between the two ZPLs well. Indeed, if the optical dipole for the V1’ transition is assumed to be nearly perpendicular to the c axis (and thus closely aligned to the cavity mode), the values of enhancement measured here would imply that the V1 optical dipole is polarized ~60° from the V1’ dipole, giving an angle of ~30° with the c axis.

In summary, we have demonstrated 1D nanobeam PCCs in 4H-SiC with high-quality factor modes (\( Q \) up to 5,300) coupled to silicon vacancy defect centers generated by ion implantation. Using high-Q cavities with resonances slightly blueshifted from two closely related silicon vacancy ZPLs, we used nitrogen gas condensation to controllably tune the cavity mode into resonance with these ZPLs. We observed Purcell enhancement of more than 80-fold and confirm these measurements by comparing on- and off-resonant intensities, increased spontaneous emission fraction into the ZPLs, and decreased defect fluorescence lifetime on-resonance with the cavity. Therefore, the increased photonic density of states when the cavity ana-

Materials and Methods

Sample Fabrication. Electron-beam lithography (Elionix) is used to define the cavity pattern, which is transferred to an aluminum hard mask by reactive ion etching (RIE). A second RIE step (STS ICP-RIE) transfers the pattern to the SiC wafer, which consists of a 250-nm p-type (Al, 5 × 10^{17} cm^{-3}) epi-layer homoepitaxially grown on a n-type substrate (N, 10^{19} cm^{-3}). After pattern transfer, the n-type substrate is removed with a dopant-selective photolithographic etch to provide optical isolation for the cavity (25, 31). This procedure allows us to create cavities out of high-quality epitaxially grown material.

Defect Implantation. Silicon vacancy centers were created by implantation of 12C ions at a dose of 10^{14} ions per square centimeter and an energy of 70 keV. In the vertical dimension, the cavity field profile reaches its maximum value in the middle of the cavity, which is 125 nm below the surface for our 250-nm device layer. Therefore, the 70-keV ion energy was selected to create defects with a maximum probability at this depth, as calculated by stopping range of ions in matter simulations.

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