

On-Demand Generation of Single Silicon Vacancy Defects in Silicon Carbide

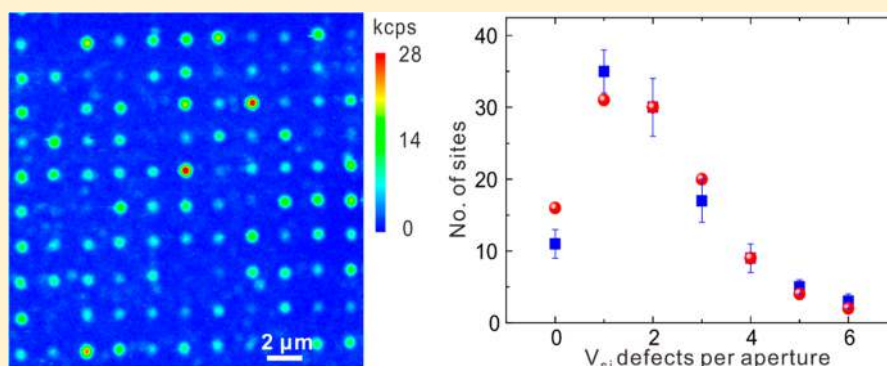
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S Supporting Information



ABSTRACT: Defects in silicon carbide have been explored as promising spin systems in quantum technologies. However, for practical quantum metrology and quantum communication, it is critical to achieve on-demand single spin-defect generation. In this work, we present the generation and characterization of shallow silicon vacancies in silicon carbide by using different implanted ions and annealing conditions. The conversion efficiency of a silicon vacancy of helium ions is shown to be higher than that by carbon and hydrogen ions in a wide implanted fluence range. Furthermore, after optimizing the annealing conditions, the conversion efficiency can be increased more than 2 times. Due to the high density of the generated ensemble defects, the sensitivity of sensing a static magnetic field can be reached as high as $\eta_B \approx 23.5 \mu\text{T}/\sqrt{\text{Hz}}$, which is about 9 times smaller than previous results. By carefully optimizing implanted conditions, we further show that a single silicon vacancy array can be generated with a high conversion efficiency of about 80%. The results pave the way for using an on-demand-generated single silicon vacancy for quantum information processing and quantum photonics.

KEYWORDS: silicon carbide, silicon vacancy, implantation, magnetic sensing, single-photon sources

In recent years, color centers in silicon carbide (SiC) have been demonstrated as promising physical platforms for quantum science.^{1–11} SiC is a well-known semiconductor material that has wide applications in high-power and high-temperature electronic devices. Moreover, SiC has technological advantages due to the well-developed device fabrication protocols and inch-scale growth. Besides some bright single-photon emitters,^{3–7} SiC also has two types of defect spins, including the silicon vacancy and divacancy defects.^{1,2,8–11} Similar to nitrogen-vacancy (NV) centers in diamond,¹² these spins can be polarized by optics and manipulated by microwaves at room temperature (RT). Moreover, their photoluminescence (PL) spectra are in the near-infrared, which would have weaker scattering losses at interfaces and

signal attenuation in optical fibers¹² than that of NV centers in diamond. Recently, silicon vacancy (V_{Si}) defects in 4H-SiC stand out as a favorable system for quantum technology including quantum information processing and quantum sensing, due to their unique properties such as photostability, half-integer $S = 3/2$ spin, and long spin coherence time at RT.^{9–11,13,14,16–21}

For practical quantum sensing and quantum communication, it is critical to generate single V_{Si} defects in 4H-SiC with high enough efficiency. Previously, there were many methods to generate V_{Si} defects: electron and neutron irradiation,^{9,11,13}

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carbon ion implantation,²⁰ focus ion beams,²¹ laser writing,^{22,23} etc. However, in order to further improve the conversion efficiencies and investigate the generation effect of V_{Si} defects, different implanted ions need to be used.^{20,21} Since the ion implantation creates residual radiation damage, it would degrade the coherence properties of V_{Si} defects to an extent that is hardly usable as a spin probe.^{2,25–27} Moreover, due to the low count of V_{Si} defects (about 10 kcps), in order to conveniently integrate with photonic devices,^{28,29} it is necessary to further improve the conversion efficiency to on-demand generation of single V_{Si} defects. Furthermore, efficiently generating high-density V_{Si} defect ensembles will tremendously increase the sensitivity in quantum sensing applications and be useful in the investigation of many-body dynamics with defect ensemble interactions.^{15–19,30,31}

In this work, we compare the generation effect of shallow V_{Si} defects in 4H-SiC by using three different implanted ions with wide implanted fluences. Through measuring the counts and optically detected magnetic resonance (ODMR) spectra, the implanted effect by helium ions is shown to be better than that by carbon and hydrogen ions in wide fluences. Moreover, we also optimized the annealing conditions for the three different implanted ions. All the conversion effects are found to rise more than 2 times at the optimal annealing conditions. The ODMR signals of the generated high-density shallow V_{Si} defect ensembles are measured with different external magnetic fields with the magnetic sensitivity detected to be about $23.5 \mu\text{T}/\sqrt{\text{Hz}}$, which is about 9 times smaller than previous results. Finally, we generated a shallow single V_{Si} defect array in SiC with a high implanted conversion efficiency of about 80%. Our on-demand generation of single and high-density ensemble of shallow V_{Si} defects would be directly used for defect-based quantum photonics and quantum sensing with technological materials.

In the experiment, we use a commercially available high-purity 4H-SiC epitaxy layer (4° off-axis, thickness is about $7 \mu\text{m}$, nitrogen doped, and concentration is $5.2 \times 10^{15} \text{ cm}^{-3}$, Xiamen Powerway advanced material) sample.^{20,21} The energy gap of the 4H-SiC is about 3.23 eV, and the negative V_{Si} defects of the Fermi level is about 1.5–2.3 eV.²⁴ In order to compare the implanted effect of different ions, we generate shallow V_{Si} defects in the sample by implanting hydrogen (H_2^+), helium (He^+), and carbon (C^+) with the same energy (40 keV for H_2^+ , 20 keV for He^+ and C^+) on the surface of the sample and with the fluence ranging from $1 \times 10^{11} \text{ cm}^{-2}$ to $1 \times 10^{14} \text{ cm}^{-2}$, respectively. For the demonstration of a generated single silicon vacancy array, first we use electron-beam lithography (EBL) to make $50 \pm 10 \text{ nm}$ diameter arrays ($2 \times 2 \mu\text{m}^2$) and $10\text{-}\mu\text{m}$ -wide strips used as position markers on a 200 nm polymethyl methacrylate (PMMA) layer, which are deposited on the SiC surface by spin coating.²⁰ Since the 20 keV helium and hydrogen implanted ions can penetrate through the 200 nm PMMA layer inferred from the stopping and range of ions in matter (SRIM) simulation,^{20,21} we chose carbon ions, which can be blocked by the PMMA layer. The sample is then implanted by the 20 keV carbon ions with a fluence of $1.3 \times 10^{11} \text{ cm}^{-2}$ to prepare the single shallow V_{Si} defect array.²⁰ The PL and spin properties of the defects are characterized by using a home-built confocal microscope combined with a microwave system.^{20,21} A 730 nm laser is used to excite the V_{Si} defects through an objective (polarization of the laser perpendicular to the quantization axis of the sample). In the RT experiment, we use three confocal setups.

For all of the scans of the defects at RT, we use a 1.3 NA oil objective (Nikon) to focus on the sample and collect the fluorescence directly by two avalanche photodiodes (APDs) with a $40\text{-}\mu\text{m}$ -diameter pinhole.^{13,20,21} For ODMR measurement, we use an infrared 0.8 NA objective (Olympus) to excite the sample, and the fluorescence is collected by a single-mode fiber to an APD. For Rabi and Ramsey measurement, we use an infrared 0.8 NA objective (Olympus) to excite the sample, and the fluorescence is collected by a multimode fiber to a Femto (OE-200-Si) (see Supporting Information). For detecting the low-temperature (LT) PL spectra, we use a Montana cryostation (4–350 K) combined with a confocal system with an infrared 0.65 NA objective (Olympus).

With the setup described, we first characterize the PL spectra and ODMR of the generated shallow V_{Si} defects by implanting hydrogen, helium, and carbon ions with the dose ranging from 1×10^{11} to $1 \times 10^{14} \text{ cm}^{-2}$ with the same energy (20 keV). Figure 1a shows the theoretical depth profiles of the

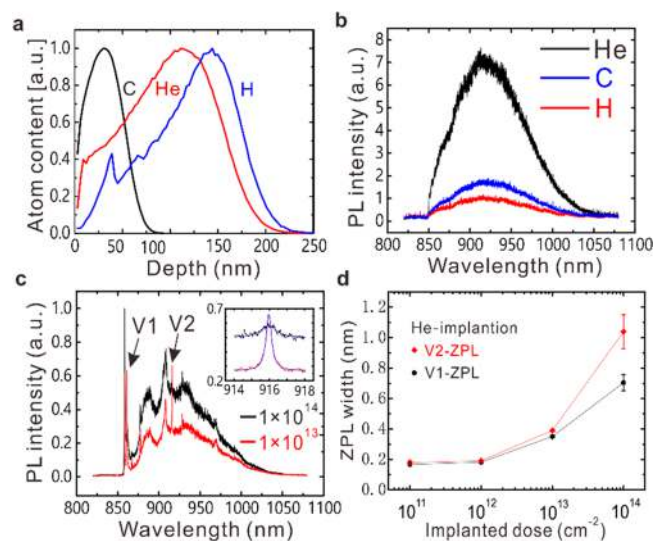


Figure 1. Characterization of the PL spectrum of the implanted shallow V_{Si} defects. (a) SRIM simulation of depth profiles of the generated shallow V_{Si} defects in 4H-SiC implanted by three types of ions with the same energy (20 keV): hydrogen (H^+), helium (He^+), and carbon (C^+). (b) Comparison of room-temperature PL spectrum of implanted V_{Si} defects by the three ions with the same fluence of $1 \times 10^{13}/\text{cm}^2$. (c) Low-temperature PL spectra of implanted V_{Si} defects by helium with a fluence of 1×10^{13} and $1 \times 10^{14} \text{ cm}^{-2}$, respectively. The inset is a zoom-in of the ZPLs of the V2 defect. The blue lines are the fits to the ZPLs data. (d) The ZPL width of V1 and V2 defects increases as the implanted dose increases.

generated shallow V_{Si} defects simulated by the SRIM.^{20,21} All the depths of the V_{Si} defects are less than 200 nm, and particularly, the depth of defects by the carbon implantation is less than 60 nm, which implies that the generated V_{Si} defects are shallow. Figure 1b shows the comparison of the RT PL spectrum of the defects generated by different implanted ions with the same dose of $1 \times 10^{13} \text{ cm}^{-2}$, which are measured by a spectrometer (iHR550, Horiba). The wavelengths of the PL spectrum ranged from 850 to 1050 nm, which is consistent with the RT PL spectrum of the V_{Si} defects as measured in previous works.^{9,13} Moreover, we can see that the sample implanted by helium ions has the highest PL intensity.

The LT (5 K) PL spectrum of the generated defects are further measured using the LT confocal system. Two

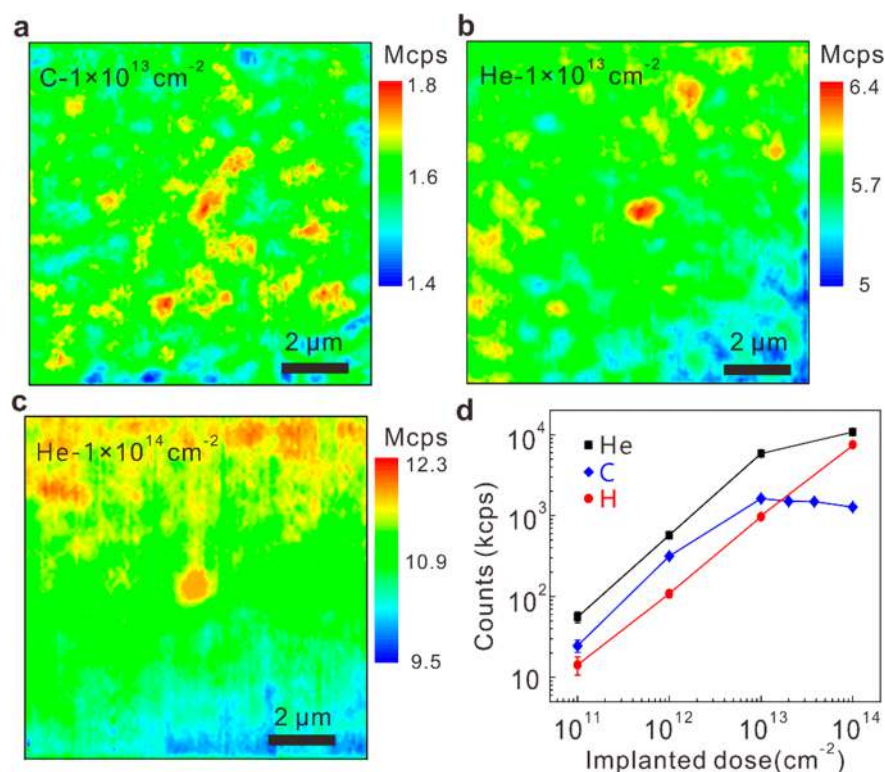


Figure 2. PL intensity of defects generated by different implanted ions for different fluences with an excitation power of 0.5 mW. Confocal microscope fluorescence images ($10 \times 10 \mu\text{m}^2$) of the implanted shallow V_{Si} defects by different implanted conditions: (a) $\text{C}-1 \times 10^{13} \text{ cm}^{-2}$, (b) $\text{He}-1 \times 10^{13} \text{ cm}^{-2}$, and (c) $\text{He}-1 \times 10^{14} \text{ cm}^{-2}$, respectively. (d) Comparison of the mean counts of the shallow V_{Si} defects generated by three different implanted ions (H_2^+ , He^+ , C^+) for different implanted doses.

representative LT PL spectra of the defects implanted by $\text{He}-1 \times 10^{13} \text{ cm}^{-2}$ and $\text{He}-1 \times 10^{14} \text{ cm}^{-2}$ are shown in Figure 1c. Two characteristic peaks at 861.3 and 916.0 nm are denoted, which are the zero phonon lines (ZPLs) of the two nonequivalent lattice sites of the V1 and V2 centers of the V_{Si} defects, respectively.^{14,20,21} Recently, the V1 center has attracted attention for its unique properties.²³ The V1 defect decouples its optical properties from magnetic and electric fields, as well as from local strain. This provides a high-fidelity spin-to-photon interface with exceptionally stable and narrow optical transitions, low inhomogeneous broadening, and a large fraction of resonantly emitted photons.^{11,32} The inset is a zoom-in of the ZPLs of the V2 defect. The blue lines are the fits to the ZPL data. The full width at half-maximum (FWHM) of these two ZPLs of the V2 defect are deduced from the fits to be 0.39 ± 0.01 and 1.04 ± 0.09 nm for the implanted doses of 1×10^{13} and $1 \times 10^{14} \text{ cm}^{-2}$, respectively. Since the ZPL width is important,^{33,34} we compare the ZPL FWHM of the generated V_{Si} defects as the implanted fluence. The results are shown in Figure 1d, and the ZPL width of V1 and V2 slightly increases as the fluence increases from $1 \times 10^{11} \text{ cm}^{-2}$ (0.18 nm) to $1 \times 10^{13} \text{ cm}^{-2}$ (0.39 nm), but then dramatically increases for the fluence of $1 \times 10^{14} \text{ cm}^{-2}$. Moreover, for all the fluences, the ZPL width of V1 is less than that of V2. The broadening of the ZPL width may stem from the increasing impurity interactions with the V_{Si} defects.³⁵

In order to compare the fluence effect of different implanted ions, we comprehensively investigate the PL intensity of the generated V_{Si} defects with an excitation power of 0.5 mW. Three representative confocal fluorescence images of the defects implanted by $\text{C}-1 \times 10^{13} \text{ cm}^{-2}$, $\text{He}-1 \times 10^{13} \text{ cm}^{-2}$, and

$\text{He}-1 \times 10^{14} \text{ cm}^{-2}$ are shown in Figure 2a, b, and c, respectively. The results show that the PL intensity of V_{Si} defects implanted by He is about 4 times higher than that of C with the same dose of $1 \times 10^{13} \text{ cm}^{-2}$. The PL intensity of V_{Si} defects implanted by He with a dose of $1 \times 10^{14} \text{ cm}^{-2}$ is about 1.7 times higher than that with a dose of $1 \times 10^{13} \text{ cm}^{-2}$.

We further compare the mean counts of the shallow V_{Si} defects generated by three different implanted ions with different implanted doses in Figure 2d. The mean counts are calculated as the average counts of the scanned $10 \times 10 \mu\text{m}^2$ areas. It can be seen that the mean counts of defects for all three implanted ions increase almost linearly with the fluence ranging from 1×10^{11} to $1 \times 10^{13} \text{ cm}^{-2}$. When the fluence is up to $1 \times 10^{14} \text{ cm}^{-2}$, the counts of hydrogen ion implantation still increase linearly, and helium ion implantation increases slowly. However, for the carbon ion implantation, the counts are shown to decrease a little, which is similar to that observed in the NV centers in diamond implanted by nitrogen ions.³⁶ The reason for the decrease might be due to the ion-induced damage of the crystal lattices, which leads to the amorphization of the SiC.³⁶ The implanted effect of helium is shown to be better than that of hydrogen and carbon for fluences ranging from 1×10^{11} to $1 \times 10^{14} \text{ cm}^{-2}$, which has the highest conversion efficiency to generated V_{Si} defects. Particularly, the mean counts of the helium implantation are about 2 and 4 times larger than that of the carbon and hydrogen implantation for a fluence of $1 \times 10^{11} \text{ cm}^{-2}$, respectively, while for the higher fluence of $1 \times 10^{14} \text{ cm}^{-2}$ the mean counts of the helium implantation are about 8 and 1.5 times larger than that of the carbon and hydrogen implantation, respectively.

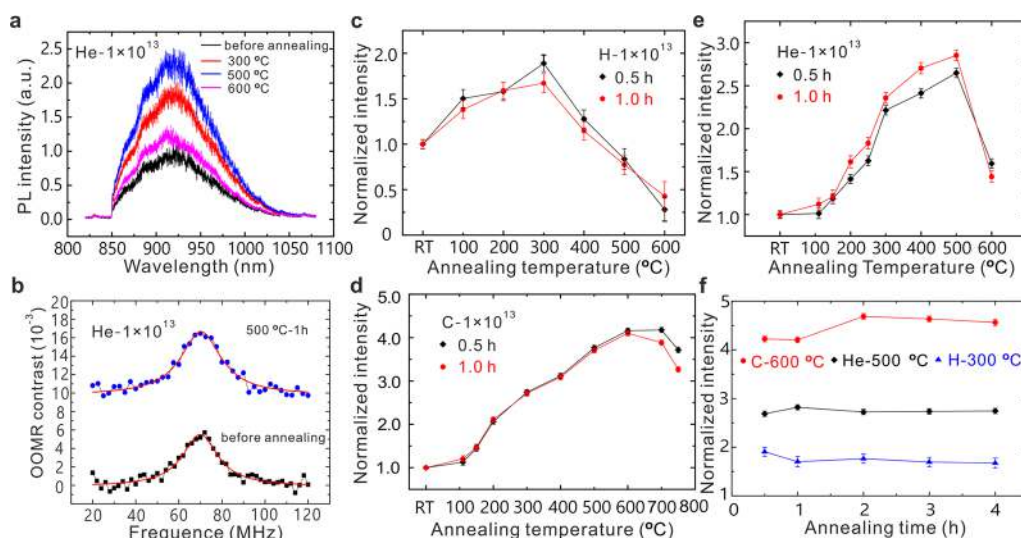


Figure 3. PL enhancement after different annealing conditions. (a) RT PL spectrum of the implanted V_{Si} defects ($He-1 \times 10^{13} \text{ cm}^{-2}$) after different annealing temperatures with an annealing time of 0.5 h. (b) ODMR measurement of the implanted shallow V_{Si} defects ($He-1 \times 10^{13} \text{ cm}^{-2}$) without annealing and after annealing at 500 °C for 1 h, respectively. The red lines are the Lorentzian fitting of the data. (c–e) Normalized intensity of the defects as a function of the annealing temperature with annealing times of 0.5 and 1 h for implanted hydrogen, helium, and carbon ions ($1 \times 10^{13} \text{ cm}^{-2}$), respectively. RT in the *x*-axis represents the case with no annealing of the sample. (f) Summary of the PL enhancement at corresponding optimized temperatures for different annealing times.

The saturation phenomenon of the V_{Si} defects generated by implanted C ions might result from the lattice damage that accumulates in the form of multivacancy defects as the diamond lattice approaches the graphitization threshold or an accumulation of charged defects in the SiC lattice that lead to ionization, similar to what was observed in NV and SiV centers in diamond.^{36–38} Since the C ion is heavier than the H and He ions, it will generate more defects. Moreover, the depth of the implanted C ions is shallower than that of H and He ions. The C ions might generate much more defects and damage than H and He ions in the same volume. So the implanted C ions' behavior is different from that of the H and He ions. In our experiment, the PL intensity is much higher than previous maximum implanted results,¹³ which would be useful for high-sensitivity quantum sensing based on V_{Si} defect ensembles and investigation of many-body dynamics with defect interactions.^{16–19,29–31}

Annealing has been shown to be an effective method to increase the density of defects,^{3,13} which is further investigated in our experiment. A glass tube furnace with a high vacuum of $5 \times 10^{-4} \text{ Pa}$ is used to anneal the sample. Figure 3a shows the PL spectrum of V_{Si} defects implanted by $He-1 \times 10^{13} \text{ cm}^{-2}$ after different annealing temperatures for 0.5 h. It is shown that the PL spectrum profiles remain the same, which implies that the annealing does not change the optical properties of the V_{Si} defects. Moreover, the PL intensity increases as the annealing temperature increases from 300 °C to 500 °C. However, it decreases after 600 °C annealing, which means that it has an optimal annealing temperature. We then study the effect of annealing on the spin property of the implanted shallow V_{Si} defects. Figure 3b shows the ODMR measurement of the V_{Si} defects ($He-1 \times 10^{13} \text{ cm}^{-2}$) at zero magnetic field without annealing and after annealing at 500 °C for 1 h, respectively. Red lines are the Lorentzian fits of the ODMR data. The ODMR spectrum is the ensemble V_{Si} defects, and the contrast of the ODMR (about 0.7%) is consistent with previous ensemble defect results.^{11,13} We find that the resonant

microwave frequencies are almost the same for the samples without annealing (69.9 MHz with a width of 20 MHz) and with annealing (70.1 MHz with a width of 22.7 MHz). All the results are consistent with previous ODMR results of the V_{Si} defects.^{9,16,20,21} Moreover we also performed a Ramsey measurement (see Supporting Information). Inferred from the fitting, the T_2^* of the preannealing sample is $130 \pm 30 \text{ ns}$ at $B = 14 \text{ G}$, which is consistent with the values of $168 \pm 10 \text{ ns}$ for the annealed sample (500 °C for 1 h). Both T_2^* values are consistent with previous results.¹⁴ This demonstrates that the annealing might have little influence on the spin properties of V_{Si} defects.

Figure 3c shows the normalized intensity of the V_{Si} defects implanted by $H-1 \times 10^{13} \text{ cm}^{-2}$ as a function of the annealing temperature with an annealing time of 0.5 h (black) and 1 h (red), respectively. The mean counts of the scanned $10 \times 10 \mu\text{m}^2$ areas before annealing are set as 1. It is shown that the normalized intensity profiles of 0.5 and 1 h annealing are almost the same, both of the optimal annealing temperatures are 300 °C, and the normalized counts increase about 2 times. As shown in Figure 3d and e, both of the normalized intensity profiles of 0.5 h (black) and 1 h (red) annealing for the $He-1 \times 10^{13} \text{ cm}^{-2}$ and $C-1 \times 10^{13} \text{ cm}^{-2}$ implantation are also almost the same, and the optimal annealing temperatures are 500 and 600 °C, respectively. Moreover, the normalized counts increase about 3 and 4 times, respectively. The difference of the optimal annealing temperatures for C, H, and He ions might result from their generating many different local types of defects. The different types of defects might play a role in the optimal annealing temperatures to generate the V_{Si} defects.^{3,13,39–42} To know the exact physical reason for the different optimal annealing temperatures, further investigation such as theory and EPR technology is needed.^{40–42} The decrease of the PL intensity after the annealing higher than 600 °C for the three implanted ions is consistent with previous results.^{3,13,43–45} We further investigated the PL enhancement effect of the annealing time at the optimal annealing temperature (C-600

°C, He-500 °C, H-300 °C), which are shown in Figure 3f. The PL enhancements are almost the same for the annealing time from 0.5 to 4 h, which demonstrates the robustness of the annealing effect.

Since the V_{Si} defect is a spin qubit and has been used in spin-based quantum information processing and quantum sensing,^{9,11,13,14,16–20,46} we further investigate the spin property of the high-concentration shallow V_{Si} defect ensembles (V2 center) (implanted by $He-1 \times 10^{13} \text{ cm}^{-2}$, annealing at 500 °C for 1 h) with an external magnetic field. The electronic ground state of the V_{Si} defects is a quartet manifold ($S = 3/2$), and its electronic spin Hamiltonian is

$$H = D[S_z^2 - S(S + 1)/3] + g\mu_B B S_z \quad (1)$$

where the zero-field-splitting (ZFS) parameter D is 35 MHz, $g = 2$ is the electron g -factor, μ_B is the Bohr magneton, and B is the applied axial static magnetic field. In zero magnetic field, the $| \pm 1/2 \rangle$ and $| \pm 3/2 \rangle$ states are both degenerate. Figure 4a

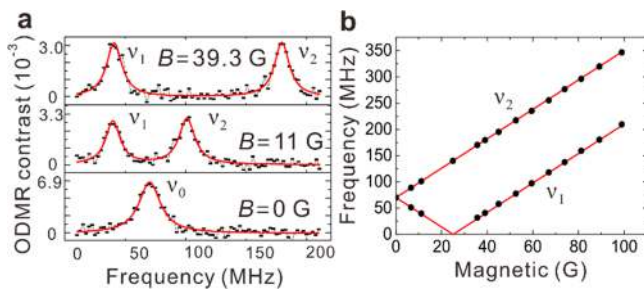


Figure 4. ODMR measurements with different external magnetic fields at room temperature. (a) ODMR measurement of the V_{Si} defects ($He-1 \times 10^{13} \text{ cm}^{-2}$, annealing at 500 °C for 1 h) for three different axial magnetic fields. The red lines are the Lorentzian fit for the ODMR data. (b) Resonance frequencies of the ODMR signal as a function of the axial magnetic field from 0 to 100 G. The red lines are the theoretical predictions using the Hamiltonian of the V_{Si} defects.

shows three ODMR measurements of different magnetic field, with the lowest panel showing the case with zero magnetic field for comparison. By increasing the external axial magnetic field B , the spin degeneracy is increased, resulting in four distinct energy eigenvalues, $E_{\pm 1/2} = -D \pm g\mu_B B/2$, $E_{\pm 3/2} = D \pm 3g\mu_B B/2$, and two dipole-allowed transitions, $| -1/2 \rangle \leftrightarrow | -3/2 \rangle$ and $| 1/2 \rangle \leftrightarrow | 3/2 \rangle$, respectively. The corresponding transition frequencies are $\nu_1 = |2D - g\mu_B B|$ and $\nu_2 = 2D + g\mu_B B$, respectively. When B is less than $2D/g\mu_B = 25 \text{ G}$, $\nu_1 = 2D - g\mu_B B$. As shown in Figure 4a (middle panel), the two resonance frequencies (width) are $\nu_1 = 39.5 \text{ MHz}$ (14.6 MHz) and $\nu_2 = 100.8 \text{ MHz}$ (15.8 MHz) with the magnetic field at about 11 G. However, when B is larger than 25 G, $\nu_1 = g\mu_B B - 2D$. As shown in Figure 4a (upper panel), the two transition frequencies (width) are $\nu_1 = 40.3 \text{ MHz}$ (15.6 MHz) and $\nu_2 = 179.9 \text{ MHz}$ (15 MHz) with the magnetic field at about 39.3 G.

The transition frequencies of the ODMR signal as a function of the axial magnetic field from 0 to 100 G are shown in Figure 4b. The red lines are the calculations for the two dipole-allowed transition frequencies using the Hamiltonian of the V_{Si} defects. The experimental ODMR spectrum is in good agreement with theoretical predictions using eq 1. To evaluate the V_{Si} defect-based magnetic sensing sensitivity, the shot-noise-limited DC magnetic field sensitivity η_B is approximated through the following equation: $\eta_B \approx \frac{h}{g\mu_B} \frac{\Delta\nu}{C\sqrt{R}}$, where h is Planck's constant, R is the rate of detected photons, C is the contrast of the ODMR, and $\Delta\nu$ is the ODMR width.^{47,48} In the experiment, the mean ODMR width $\Delta\nu$ is 15.3 MHz, the contrast of the ODMR C is about 0.3%, and the maximum detected count of the V_{Si} defects R is about 60 Mcps, which corresponds to 5×10^3 defects in the about $0.4 \mu\text{m}^2$ in the xy plane and $1 \mu\text{m}$ in the z -direction sensing volume.⁴⁸ We can then deduce the sensitivity $\eta_B \approx 23.5 \mu\text{T}/\sqrt{\text{Hz}}$, which is about 9 times smaller than previous results.¹⁷ It can be

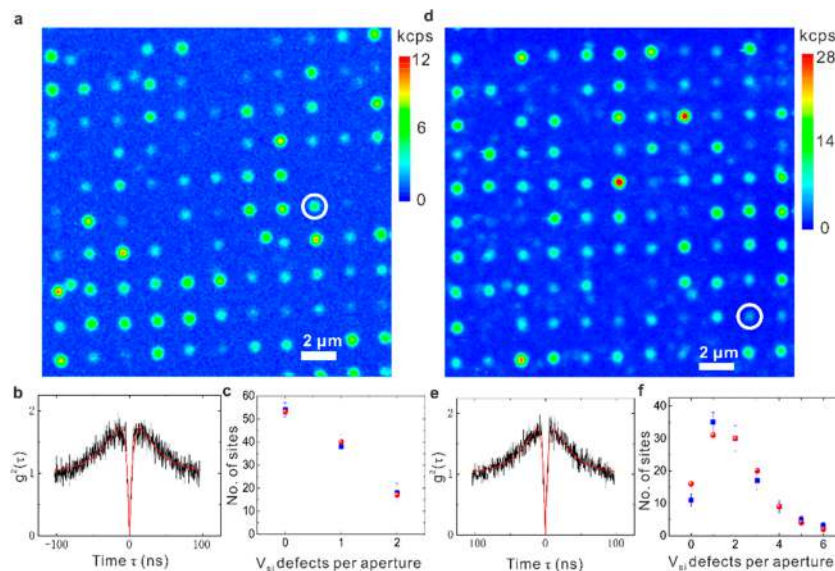


Figure 5. Characterization of the carbon-implanted shallow single V_{Si} defect arrays. (a and d) Two representative confocal fluorescence images ($20 \times 20 \mu\text{m}^2$) of the implanted shallow single V_{Si} defect arrays before and after annealing (600 °C for 1 h) with an excitation power of 0.5 mW, respectively. The scale bars are $2 \mu\text{m}$. (b and e) Second-order correlation function measurement of the single V_{Si} defects (corresponding to the circled single V_{Si} defects in (a) and (d), respectively). The red lines are the fits of the data. (c and f) Statistics of the number of V_{Si} defects per implanted aperture (blue points). The data are fitted with the Poisson distribution (red points).

expected that the ODMR width can be further reduced to 0.5 MHz using the pulsed ODMR, and the corresponding magnetic sensitivity can be further improved to about 770 nT/ $\sqrt{\text{Hz}}$.¹⁷ Moreover, the magnetic sensitivity can be further improved by using the isotopically purified SiC substrate.⁴⁹

Finally, we characterize the generated efficiency of the carbon-implanted shallow single V_{Si} defect arrays. As a contrast, we investigated two samples: one is as implanted, and the other is optimal annealing (600 °C for 1 h). Figure 5a and d are two representative confocal fluorescence images ($20 \times 20 \mu\text{m}^2$) of the implanted shallow single V_{Si} defect arrays before and after annealing with an excitation power of 0.5 mW, respectively. It was shown that in both conditions they display clear arrays with a prominent increase of the concentration in the annealed sample. In order to identify a single V_{Si} defect, we measure the corresponding second-order correlation function of the defect in the implanted aperture denoted by the white circle in Figure 5a and d, respectively. Since the fluorescent count rate of the single V_{Si} defect is low (maximum counts is about 10 kcps), the background fluorescence has been included in the evaluation of $g^2(\tau)$. The autocorrelation function $g^2(t)$ is corrected from normalized raw data $C_N(t)$ using the function $g^2(\tau) = (C_N(\tau) - (1 - \rho^2))/\rho^2$, where $\rho = s/(s + b)$, where s and b are the signal and background counts, respectively.^{8,13,20,21} The background-corrected $g^2(\tau)$ are shown in Figure 5b and e, respectively. The red lines are the fits according to the function $g^2(\tau) = 1 - (1 + a) \exp(-|\tau|/\tau_1) + a \exp(-|\tau|/\tau_2)$, where a , τ_1 , and τ_2 are fitting parameters. It can be seen that, in both cases, $g^2(0)$ is close to 0, which demonstrates they are single V_{Si} defects.

Then we count the number of V_{Si} defects per implanted aperture for 110 apertures in both samples, and the corresponding results are shown in Figure 5c and f, respectively. The data (blue points) are fitted using Poisson distribution (red points). Inferred from the fits, the average number of V_{Si} defects formed per aperture is 0.75 ± 0.05 and 1.94 ± 0.12 , respectively. Since the implanted fluence corresponds to about 2.5 carbon atoms per aperture, the conversion yields of the implanted carbon ions into the V_{Si} defects are about $30\% \pm 2\%$ and $78\% \pm 5\%$, respectively. The high conversion yield (about 80%) is about 4 times higher than previous results,²⁰ which reaches the highest conversion yield in solid state systems.⁵⁰

In summary, we provide a method to on-demand generate shallow single V_{Si} defect arrays with a high conversion efficiency of about 80% and high concentration defect ensembles in 4H-SiC. The comparison of implanted results demonstrate that the effects of helium implantation are better than that of hydrogen and carbon implantation from a low fluence ($1 \times 10^{11} \text{ cm}^{-2}$) to a high fluence ($1 \times 10^{14} \text{ cm}^{-2}$). Through optimizing annealing temperature and annealing time, the PL intensity increases about more than 2 times. Moreover, using the implanted high concentration ensemble V_{Si} defects, the magnetic sensing sensitivity can reach $\eta_B \approx 23.5 \mu\text{T}/\sqrt{\text{Hz}}$, which is about 9 times smaller than previous results.¹⁷ The magnetic sensitivity can be further improved by using pulsed ODMR¹⁷ and the isotopically purified SiC substrate.⁴⁹

Our experimental results open up several practical applications. First, the high conversion efficiency of single V_{Si} defects would lead to little residual radiation damage,²⁵ which can prolong the coherence time of defect spins and is useful in

quantum information processing¹⁴ and quantum sensing.^{16,17} Moreover, highly efficient generation of V_{Si} defects will also be good for coupling V_{Si} defects to photonics devices.^{9,28,29} Second, the high concentration V_{Si} defect ensembles can be used to achieve highly sensitive magnetic and temperature sensing^{16–19,51–53} and to study many-body dynamics with defect interactions.^{30,31} Using laser ablation⁵⁴ or reactive ion etching,⁵⁵ we can make high-quality nano-SiC particles with the implanted sample, which would lead to high sensitivity, nanoscale spatial resolution sensors with photostability and chemical inertness. Finally, the methods may be used to generate other types of defects in SiC with high quality, such as divacancies.^{1,2}

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.9b00451.

PL image of a single defect and its saturation curve; Rabi and Ramsey measurement of the $\text{He-1} \times 10^{13}$ sample before and after annealing at 500 °C for 1 h (PDF)

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Notes

The authors declare no competing financial interest.

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