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Efficient Generation of an Array of Single Silicon-Vacancy Defects in Silicon Carbide

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Color centers in silicon carbide have increasingly attracted attention in recent years owing to their excellent properties such as single-photon emission, good photostability, and long spin-coherence time even at room temperature. As compared to diamond, which is widely used for hosting nitrogen-vacancy centers, silicon carbide has an advantage in terms of large-scale, high-quality, and low-cost growth, as well as an advanced fabrication technique in optoelectronics, leading to prospects for large-scale quantum engineering. In this paper, we report an experimental demonstration of the generation of a single-photon-emitter array through ion implantation. $V_{\text{Si}}$ defects are generated in predetermined locations with high generation efficiency (approximately 19% ± 4%). The single emitter probability reaches approximately 34% ± 4% when the ion-implantation dose is properly set. This method serves as a critical step in integrating single $V_{\text{Si}}$ defect emitters with photonic structures, which, in turn, can improve the emission and collection efficiency of $V_{\text{Si}}$ defects when they are used in a spin photonic quantum network. On the other hand, the defects are shallow, and they are generated about 40 nm below the surface which can serve as a critical resource in quantum-sensing applications.

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I. INTRODUCTION

Silicon carbide (SiC) stands out for its application in quantum science in recent years due to its outstanding features. The main driving force behind exploring SiC comes from its advantage in chip integration over diamond, another material which is used widely to host a nitrogen-vacancy center. The same as diamond, SiC is a group-IV material, making it a nuclear-spinless environment to protect the coherence of color-center electron spins. Compared to diamond, the unique property of SiC includes high-quality, large wafer-scale growth, and mature nanofabrication and doping techniques [1–17]. SiC has been explored to generate single-photon emission in the visible [7,8] as well as near-infrared range [4,6]. Thanks to the possibility of SiC-doping control, an electrically driven SiC single-photon emitter has been created with the fabrication of emitting diodes [12]. In addition to its photonic properties similar to the nitrogen-vacancy centers in diamond [18], silicon-vacancy ($V_{\text{Si}}$) and -divacancy defects can be used as spin-qubit carriers and can be optically polarized and controlled by microwaves [1–3,5,6,9,10,13]. For $V_{\text{Si}}$ defects, the coherent control of a single spin has been achieved at room temperature, and the coherence time reaches about 160 μs [3]. Moreover, the coherence time of the $V_{\text{Si}}$ defect ensemble has been improved to about 20 ms using dynamical decoupling at cryogenic temperature [15]. In particular, the $V_{\text{Si}}$ defect has also been shown to be promising for the quantum sensing of magnetic field [9,11] and temperature [9].

For its application in quantum-information science, efficient generation of a single defect and single-photon source in SiC are required. Recently, two methods have been developed to generate single $V_{\text{Si}}$ defects: high-energy electron irradiation [3] and neutron irradiation [4]. However, in both methods, the generation efficiency of the single $V_{\text{Si}}$ defects is low, and the positions of the single $V_{\text{Si}}$ defects are randomly distributed. For application in a spin photonics network [19], the creation of defects in well-defined locations is essential in order to enhance their emission efficiency by integrating them into photonic structures.

In this work, we experimentally realize the efficient generation of a nanoscale single $V_{\text{Si}}$-defect array in SiC. The $V_{\text{Si}}$-defect array is created by using 30-keV carbon ion implantation through an array of (65 ± 10)-nm-diameter
apertures patterned on a PMMA layer using electron-beam lithography (EBL) deposited on top of the SiC surface [20]. We first measure the photoluminescence (PL) spectrum and second-order autocorrelation function \( g^2(t) \) of the defect emission at room temperature and confirm that they are single defects. We then study their saturation behavior and photostability, showing that the fluorescence emission is very stable without any indication of photoblinking. By sampling the \( V_{Si} \) defects on 100 implanted sites, we estimate that the efficiency of single silicon-vacancy defect generation is about \( 34\% \) and the conversion yield of the implanted carbon ions into the \( V_{Si} \) defects is about \( 19\% \). Finally, to confirm their origin further, we measure the PL spectrum and the optically detected magnetic resonance (ODMR) signal of the \( V_{Si} \) defects at both room temperature and low temperature (5 K).

II. EXPERIMENT

We first introduce the production steps for the defect centers. We start with a commercially available high-purity 4H-SiC epitaxy layer (its thickness is about 7 \( \mu m \)) sample. This ensures a low background suitable for single defect generation [3,4]. As shown in Fig. 1(a), after cleaning with acetone and isopropanol (IPA) in an ultrasonic bath, a 300-nm-thick PMMA layer (\( A_7 : A_4 = 5 : 3 \)) is deposited on the SiC surface by spin coating [20–22]. In the second step, an array of apertures with a 2-\( \mu m \) cell separation and some long 10-\( \mu m \)-wide strips used as position marks are generated using EBL [20]. After the development step, a series of holes are generated on the PMMA layer. With a scanning electron microscope (SEM), we can clearly see a hole about 65\( \pm \)10 nm in diameter [Figs. 1(a) and 1(b)]. Afterwards, 30-keV C\(^+\) ions are implanted with a fluence equal to \( 2.6 \times 10^{11} \text{C}^+/\text{cm}^2 \) to generate the \( V_{Si}\)-defect array. The fluence corresponds to an average of around 8.6 carbons per aperture. Using SRIM (stopping and range of ions in matter) software simulation, we find that more than 99\% of the 30-keV carbon atoms can be blocked by the PMMA layer, such that the defect can be implanted only through the apertures. After the implantation, the PMMA layer on the sample is removed by ultrasonication in acetone. Finally, the sample is cleaned by ultrasonication in IPA. In order to avoid the generation of other types of PL defects, the sample is not annealed [3]. Different from the generation of N-V center in diamond, the absence of annealing steps largely simplifies the production procedure. In order to estimate the accuracy of the \( V_{Si}\)-defect position, we consider the ion-straggling effect, simulated by SRIM software simulation. The average depth of the \( V_{Si}\) defects is about 42 nm, and the longitudinal straggling is about 35 nm.

III. RESULTS AND DISCUSSION

After the production process, we characterize the emission properties of the defects. First, we study the PL property of the emitters in a homemade confocal microscopy system [4]. A 690-nm continuous-wave laser is used to excite the \( V_{Si} \) defects through a high-N.A. (1.3) oil objective lens (Nikon). The fluorescent photons from the \( V_{Si} \) defects are collected by the same objective lens and transmitted through a dichroic mirror (801 nm). In order to suppress the background, the fluorescent photons are passed through a 75-\( \mu m \)-diameter pinhole between two lenses followed by a 900-nm long-pass filter. The photons are then directly detected by two avalanche photodiodes after a beam splitter in a Hanbury Brown and Twiss (HBT)
FIG. 2. (a) Second-order correlation function measurement with excitation power of 0.5 and 2 mW, respectively. The blue lines show the fitting of the data with the function introduced in the main text. (b) The PL count curve of the single \( V_{\text{Si-}} \) defect as a function of the excitation power. (c) The PL count of the single \( V_{\text{Si-}} \) defect as a function of time showing the photostability. The time bin is 100 ms and the excitation power is 2 mW here. (d) The histogram of the number of \( V_{\text{Si-}} \) defects per aperture. The red line is the fit of the data by a Poisson distribution function.
spectrum. The $V_{Si}$ defect is a point defect consisting of a silicon vacancy associated with four adjacent carbons in 4H-SiC. There are two types of $V_{Si}$ defects in 4H-SiC: $V_1$ and $V_2$ centers. At cryogenic temperature, there are two zero-phonon lines (ZPLs), 861.4 and 916.3 nm, corresponding to the $V_1$ and $V_2$ centers, respectively [4,16]. A spectrum of a defect ensemble is shown in Fig. 3(b), from which we can see clearly the $V_1$ and $V_2$ peaks.

Last, we measure the continuous-wave ODMR for the implanted $V_{Si}$ defects. Figure 3(c) shows a 20-μm-wide microwave stripline fabricated by standard EBL technology. Only the ODMR of the $V_2$ center is studied in this work. Its negatively charged ground state is a spin quartet state with $S = \frac{3}{2}$ exhibiting a zero-field splitting, and $D = 35$ MHz, which can be polarized by a laser and controlled by microwave [3,4,16]. Figures 3(d) and 3(e) show the ODMR measurement of the ensemble $V_{Si}$ defects at cryogenic temperature and room temperature, respectively. According to the Lorentz fit of the data, we obtain the resonant frequency as 68.4 MHz with its linewidth (FWHM) 15.7 ± 1.8 MHz for low cryogenic temperature. For room temperature, the resonant frequency is at 65.1 MHz, and its linewidth (FWHM) is 25.0 ± 1.4 MHz, which is consistent with previous results [4,11]. The slight difference might come from residual strains in different samples, and the linewidth broadening can be affected by the decoherence of the spin qubits in the nonannealed implantation sample.

**IV. SUMMARY**

In summary, we experimentally demonstrate the generation of a single silicon-vacancy defect array in silicon carbide with high efficiency. The successful generation of a single silicon-vacancy defect in a well-defined position around tens of nanometers may open up several immediate research possibilities. First, a critical factor in quantum magnetometry with a color center is the closeness of the sensing defect to the surface [27]. This is because the dipolar magnetic fields decay as the third inverse power of the distance between the sensing spin and magnetic field target. We, therefore, need to have the defect centers close enough to the sample surface. The created shallow defects may find their application in nanoscale magnetometry, especially if it is combined with chemical etching [28–30]. Second, it might serve as an efficient way to engineer spin-spin entanglement through dipole-dipole coupling [31–33]. Again, because of the coupling decays as the distance between the two defect spins with $d^{-3}$, it is much more favorable to generate two defects close to each other in the production procedure. The possibility to generate a few defects in the tens of nanometers scale, as we demonstrate here, can be used towards this direction. Third, to precisely couple the single emitter to a photonic crystal cavity [12,34–36], solid immersion lenses [37], or nanopillar structures [38,39], one needs a precise location with accuracy in the order of tens of nanometers. The method we demonstrate here may ease the coupling of the emitter to the fabricated cavity or waveguide [34–36]. Finally, with modified parameters and possibly additional, annealing steps, similar production of other types of single defects, such as divacancies [1,2,6] and carbon antisite-vacancy pair defects [8] may become possible in future.

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Montana cryostation and then fed to a microwave stripline. After it is amplified by the amplifier, it is sent into a signal is generated by signal generator and then gated by a objective is used to excite the defects. The microwave (M4081441) and the Astar QTE Project.

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repeated 20,000 times[16]. The photon counts in each on and wave signal is gated on and off with 2.8-ms duration and results are averaged. For one point in each scan, the micro-

40 to 100 MHz are conducted six times, and then the scan To decrease the fluctuation noise, ODMR scans from on the sample surface by the standard lithography method.

The whole experiment is synchronized by pulse blaster. The final ODMR contrast is calculated by \( \Delta P_L = \frac{\Sigma N(on) - \Sigma N(off)}{\Sigma N(off)} \).


