PHOTONICS Research

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Spin defects in silicon carbide (SiC) have attracted much attentions in various quantum technologies. In this work, we study the optical manipulation of charge state and coherent control of multifold divacancy spins ensemble in SiC under resonant excitation. The results reveal that the resonantly excited divacancy ensemble counts have dozens of enhancements by repumping a higher-energy laser. Moreover, it has a similar optimal repump laser wavelength of around 670 nm for multiple divacancies. On the basis of this, the optically detected magnetic resonance (ODMR) experiment shows that repump lasers with different wavelengths do not affect the ODMR contrast and line width. In addition, the repump lasers also do not change the divacancy spins' coherence times. The experiments pave the way for using the optimal repump excitation method for SiC-based quantum information processing and quantum sensing. © 2021 Chinese Laser Press

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1. INTRODUCTION

In recent years, color centers in silicon carbide (SiC) have emerged as key systems for quantum sensing and quantum information processing [1-20]. The host material of SiC is a semiconductor with mature growth technology and has wide applications in various electronic devices [1-6]. Some color centers in SiC with single-photon emission at visible and infrared wavelengths have been investigated and been used for quantum photonics [3,4]. Thanks to the near-infrared photoluminescence (PL) and long spin coherence time, optically active spin defects such as silicon vacancy (V_{Si}) [5,7], divacancy (VV) [1,2,6,8,9], and nitrogen-vacancy (NV) centers [10,11] in SiC have been extensively studied and been used in various quantum technologies. The divacancy is composed of a silicon vacancy adjacent to a carbon vacancy, which exists in 4H, 6H, and 3C SiC polytypes [1,2,6,8,9]. For example, in 4H-SiC, there are seven types of divacancy, including PL1 to PL7 [1,2,6]. Coherent control of single divacancy in 4H-SiC and 3C-SiC with long coherence time and high optically detected magnetic resonance (ODMR) readout contrast even at room temperature has been revealed [6,9,12]. The divacancy has been widely applied in quantum photonics [13], highfidelity quantum register [14], spin photon interface [9], hybrid spin-mechanical systems [15], and various kinds of high-sensitivity quantum sensing, including electric field [16], magnetic field [17], temperature [18,19], strain [20], and so on.

Similar to NV centers (NV⁻ and NV⁰) in diamond [21], the divacancy in 4H-SiC also has a bright charge state (neutral spin state VV^0) and a dark charge state (VV^-) [22–25]. Understanding the charge state is vital to the applications in quantum technologies [24,25]. Previous experiments have shown that the bright VV⁰ converts to the dark VV⁻ excited by the off-resonant laser with a wavelength larger than 980 nm [22-24]. Moreover, the divacancy PL spectra can be enhanced about 50 times by adding a repump ultraviolet laser under an off-resonant excited laser [24]. However, the specific divacancy enhanced PL spectra may be mixed by the phonon sideband of four types of divacancy PL1-PL4 due to the off-resonant excitation. On the other hand, the optimal repump laser wavelength is around 710 nm for PL2 divacancy using resonant excitation [25]. However, whether the other types of divacancy have the same optimal repump laser wavelength and whether the optimal repump laser will affect the spin property of the divacancy still need to be studied, as they are important for

divacancy-based high-sensitivity quantum sensing and quantum information processing.

To solve this problem, here we investigate the optimal repump laser wavelength for multiple divacancies and its effect on the spin property of divacancies ensemble in 4H-SiC under resonant excitation. First, we measure the high resolution resonantly excited PL intensity of multiple divacancies with and without repump lasers at low temperature. On the basis of this, PL intensity as a function of the repump laser wavelength is performed to confirm the optimal repump laser wavelength. Furthermore, ODMR experiments show that the ODMR signal contrast and line width are independent of the repump laser wavelength, power, and resonant excited laser power. Finally, we compare the coherence time of multiple divacancies between the optimal repump excitation and the off-resonant excitation and find the coherence time is almost the same. Our work will be helpful for optimal repump excitation applications in divacancy-based quantum technologies.

2. EXPERIMENTS AND RESULTS

In this work, a low-temperature home-built confocal system equipped with a cryostation (Montana) is used [17–19]. Two tunable resonant lasers (Toptica) are used for the photoluminescence excitation (PLE) resonant scan of each divacancy in 4H-SiC. We use different wavelengths of lasers for repump excitation. Different phonon sideband (PSB) ranges are detected for different divacancies. The detected PSB ranges of PL1 and PL2 are larger than 1150 nm, while for PL4, the detected PSB range is larger than 1100 nm [11]. A superconducting single-photon detector is used to collect the PSB fluorescence through a single-mode fiber [19]. A 20 µm copper wire is used to transmit the microwave to control the divacancy spin. For the ODMR experiment, we apply the lock-in method with a photoreceiver (Femto, OE-200-IN1) [17]. A Helmholtz coil is used in the experiment to generate the *c*-axis magnetic field. The experiments are performed using a high-purity semiinsulating (HPSI) 4H-SiC (Cree) sample, and the sample temperature is kept at around 35 K.

In order to check the PLE scan, we first measure the PL spectrum of the divacancies in 4H-SiC [Fig. 1(a)]. Each divacancy's zero-phonon line (ZPL) emission spectrum is consistent with previous divacancy ZPLs results [1,2]. Similar to previous experiments, when the exciting wavelength is higher than about 980 nm, the VV will convert to dark state VV⁻ through hole emission to the valence band by a two-photon process [8,24]. To investigate the ZPL absorption spectra of multiple divacancies, we perform the PLE resonant scan (keeping the same laser power) [11,26,27]. As shown in Fig. 1(b) (red hollow squares), the counts are small due to the ionization to the dark state VV⁻, which is the same as previous results [8]. Moreover, when the pumping tunable laser (100 μ W) is resonant with the ZPL absorption peak of PL1, the PLE counts will have an apparent resonant peak, which is similar to the NV centers in SiC [11,26,27]. Moreover, inferred from the fit, the absorption peak of PL1 is 1132.17 ± 0.01 nm, which is consistent with the measured ZPL emission peak of 1132.20 nm [11].

Furthermore, previous research has shown that introducing a high-energy repump laser (near-ultraviolet) can enhance the PL intensity of the PL1–PL4 dozens of times by returning the negative charged VV⁻ divacancy to a bright state VV⁰ under an off-resonant laser of 976 nm [24]. Given this, we introduce a 532 nm (10 μ W) repump laser for the PLE resonant scan [red solid squares in Fig. 1(b)]. The repump laser does not



Fig. 1. (a) Low-temperature PL spectrum of the divacancies in 4H-SiC. Low-temperature PLE resonant scan for divacancy (b) PL1, (c) PL2, (d) PL4, (e) PL5, and (f) PL6. The solid and hollow symbols are the experimental results under repump and pure PLE excitation, respectively. The PLE spectra are fit using Lorentz functions.

change the ZPL absorption peak and enhances the PLE counts about 40 times due to the charge reset effect of the 532 nm laser [8,24]. Similarly, two different repump lasers of 730 and 532 nm (10 μ W) also enhance the PLE resonant scan intensity for PL2 [Fig. 1(c)] and PL4 [Fig. 1(d)] for around 27 and 40 times, respectively. The enhancement of the PLE counts under resonant excitation is similar to previous results under off-resonant excitation [22–24]. We also perform the PLE resonant scan of PL4 under different repump laser wavelengths. Different wavelengths of repump lasers do not affect the ZPL absorption peak. Besides, the 532 nm repump laser has a similar enhancement with the 730 nm laser, both enhancements of which are much larger than that of 920 nm laser. The result demonstrates that different wavelengths of repump lasers have different PLE enhancement effects.

Recently, Ivady *et al.* showed that the PL5 and PL6 centers are specific divacancies in stacking-fault structures, which have different natures compared to the PL1–PL4 centers [28]. In view of this, we also measure the PLE scan of PL5 [Fig. 1(e)] and PL6 [Fig. 1(f)]. The measured PLE peaks of PL5 (1042.41 nm) and PL6 (1137.96 nm) are consistent with the corresponding measured ZPLs at 1042.2 nm (PL5) and 1137.8 nm (PL6). The quantum well defined by a stacking fault can stabilize the charge states of PL5 and PL6 [24,28].

Then we investigate the impact of the different repump lasers on the PLE intensity for each divacancy. The PLE counts (solid squares) and the corresponding PL enhancement (hollow squares, the ratio between the PLE intensity with the repump laser and without the repump laser) of PL1 as a function of the repump laser wavelength are shown in Fig. 2(a). The PL counts have a small increase as the repump laser wavelength increases from 400 to 670 nm, and then they have a rapid decrease as the repump laser wavelength increases to 920 nm. The optimal repump wavelength is around 670 nm [8]. Similarly, the PLE counts and corresponding PL enhancement of PL2 and PL4 as a function of repump laser wavelength are presented in Figs. 2(b) and 2(c), respectively. Both PL2 and PL4 have a similar optimal repump wavelength at around 670 nm, which is consistent with PL1. The results demonstrate that each type of divacancy has the same optimal repump wavelength around 670 nm, which is consistent with previous results of single PL2 divacancy, indicating a possible defect absorption resonance or a specific trap-state energy [8].

Then we investigate the optical property, PLE resonant counts. The PLE counts saturation curve of PL1 with respect to the repump power is displayed in Fig. 2(d). We fit the data using a fitting function $I(P) = I_s/(1 + P_0/P)$, where P_0 is the saturation power and I_s is the maximum count. The inferred saturation power P_{0} is around 0.61 \pm 0.06 $\mu W,$ demonstrating that a tiny repump laser can retune the divacancy to a bright state. Similar ensemble results are also observed in off-resonant excitation [22-24]. Simultaneously, we also measure the PLE counts as a function of optimal pump power. In Fig. 2(e), the result shows that saturation power P_0 is around $67.4 \pm 6.4 \ \mu\text{W}$. The PL2 and PL4 divacancies also have similar saturation behaviors. Similar results have been observed by using off-resonant pump lasers [24] and single divacancy [8]. In order to check the robustness of the repump excitation, we measure the PLE resonant counts of PL1 trace at the 0.1 s time bin for three different repump laser wavelengths. All the counts remain constant. Moreover, the PLE count under repump laser of 670 nm is larger than that of 808 nm and 920 nm. These experiments show that different divacancies have similar optimal repump excited wavelengths



Fig. 2. PLE resonant counts (solid symbols) and the corresponding PL enhancement (hollow symbols) as a function of the repump laser wavelength for divacancy (a) PL1, (b) PL2, and (c) PL4, respectively. (d) and (e) PLE resonant counts of PL1 as a function of the repump and pump laser power, respectively. (f) PLE resonant counts of PL1 trace at the 0.1 s time bin using three different repump laser wavelengths.

and similar power saturation curves, even though they have different ZPLs.

The spin properties are the cornerstone of the divacancybased quantum technology applications [1,2,6,8,9]. ODMR is vital for quantum-sensing sensitivity such as DC magnetic field [16] and electric field [17] sensing and broad temperature sensing [18,19]. Given this, we then study the dependence of divacancy ODMR signals on the repump laser. The ODMR signals are detected by the standard lock-in methods [1,2,18,29,30]. The ODMR contrast is defined as $\Delta PL/PL =$ $V_{\rm Mod}/V_{\rm Tot}$, where $V_{\rm Mod}$ is the magnitude of the 70 Hz modulated component of the output voltage, and V_{Tot} is the total time-averaged output voltage [1,30]. Figure 3(a) compares the ODMR spectrum of PL1 at the zero magnetic field with the pure resonant excitation (1 mW) and various repump lasers (5 μ W). Inferred from the fits, all the ODMR resonant frequencies are around 1336.9 MHz, which indicates that repump lasers do not affect the ODMR resonant frequency. For the optimal repump wavelength (670 nm), both the V_{Mod} and V_{Tot} are enhanced around 68 times. Figure 3(b) shows the OMDR contrast and the full width at half-maximum (FWHM) as a function of repump laser wavelength. Both the ODMR contrast and FWHM are independent of the repump laser wavelength.

Similar results are also observed in the PL2 and PL4 divacancies. Furthermore, the ODMR spectrum as a function of the optimal repump laser (670 nm) power is presented in Fig. 3(c). We can see that both the OMDR contrast and

the FWHM remain almost the same as the repump laser power increases. As shown in Fig. 3(d), the pump laser power also does not affect the OMDR contrast and the FWHM. The experiment demonstrates that the resonant excitation with a repump laser does not affect the spin state and the spin readout mechanism, which is the same for the off-resonant excitation [24]. It can simultaneously enhance the PL counts dozens of times and maintain the ODMR contrast and line width, which is vital for high-sensitivity quantum sensing. For example, the divacancy DC magnetic field sensitivity is $\eta_B \approx \frac{h}{g\mu_B} \frac{\Delta \nu}{C\sqrt{R}}$, where h is the Planck constant, R is the rate of detected photons, C is the contrast of the ODMR, and $\Delta \nu$ is the ODMR width [17]. Since the PL count R in the ODMR measurement is enhanced around 68 times and the contrast and line width remain constant, the magnetic field sensitivity can have an 8.2 times enhancement for PL1 divacancies.

Spin coherence properties are the foundation for the SiCbased quantum information processing and quantum network. To this end, we investigate the impact of the repump laser on the spin coherence times of different divacancies. Figures 4(a) and 4(b) show the measurements of Ramsey and spin echo for PL2 under resonant laser (with optimal 670 nm repump laser) and off-resonant (920 nm) laser, respectively. Similar to the ODMR results, the repump laser does not affect the signal contrast. The collapses and revives in spin echo measurements are due to the Larmor frequencies of ²⁹Si and ¹³C nuclei at the corresponding magnetic field. For the Ramsey oscillations, we fit the data using the function of $I = A \cdot \exp[-(t/T_2^*)^n]$.



Fig. 3. (a) PL1 ODMR signals under PLE excitation with different repump laser wavelengths (5 μ W) at the zero magnetic field. (b), (c), and (d) The ODMR contrast and FWHM as a function of repump laser wavelength, laser power (670 nm), and resonant pump laser power, respectively. All the black squares in (b)–(d) are the data with the pure resonant pump laser.



Fig. 4. (a) and (b) Measurements of the Ramsey and spin echo of the PL2 under resonant laser (with optimal 670 nm repump laser) and off-resonant (920 nm) laser at a magnetic field of 46 G, respectively. The lines are the fits to the data. (c) Comparison of the dephasing time T_2^* and coherence time T_2 under the resonant laser (solid symbols) and off-resonant laser (hollow symbols) for PL1 (at a magnetic field of 46 G), PL2 (at a magnetic field of 46 G), and PL4 (at zero magnetic field), respectively.

 $\cos(2\pi f t + \varphi) + B$, where A, n, φ , and B are fitting parameters, and T_2^* is the spin dephasing time [18]. For the fitting of spin echo, a function $f(t) = a + b \cdot \exp[-(t/T_2)^2] \cdot [1 - t]$ $k_1 \sin^2(\pi \nu_1 t) [[1 - k_2 \sin^2(\pi \nu_2 t)]]$ is used [1,10,17], where a, b, k_1 , and k_2 are fitting parameters; ν_1 and ν_2 are related to the Larmor frequencies expected for ¹³Si and ²⁹C nuclei at the corresponding magnetic field; and T_2 is the coherence time. Figure 4(c) (middle panel) shows the fitted dephasing time T_2^* and coherence time T_2 under the resonant laser and off-resonant laser for PL2, respectively. It shows that both the T_2^* and T_2 under resonant laser are consistent with that of the off-resonant excitation. Similar results are also found for PL1 and PL4 divacancies [the left and right panels in Fig. 4(c), respectively]. The experiments show that the resonant laser does not affect the divacancy spin coherence properties, which is the same for the off-resonant excitation. The robustness of the ODMR signal and spin coherence time under resonant excitation with repump lasers are important for applications in quantum information technologies.

3. CONCLUSION

In conclusion, we study the effect on the multiple divacancy ensemble PLE counts and spin property of resonant excitation with repump lasers in 4H-SiC. The results show that the multiple divacancies have the same optimal repump laser wavelength of around 670 nm, which can robustly enhance the PLE counts dozens of times, and the repump laser does not change the PLE resonant peaks. The ODMR contrast and FWHM are also found to be independent of the repump laser wavelength and power and the pump laser power. The increased photon counts can enhance the magnetic field sensing sensitivity a few times. Finally, we demonstrate that repump laser also does not change divacancy spin coherence time. The techniques presented may also be used for silicon vacancy [5,30,31] and NV centers [10,11] in SiC. The experiment constitutes a basis for the optimal repump laser technology applications in SiC-based high sensitivity quantum sensing, quantum information processing, and quantum networks.

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