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SINGAPORE

**SINGLE-PHOTON SOURCES IN DIAMOND, SILICON CARBIDE AND
GALLIUM NITRIDE**

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SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

2018

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SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

A thesis submitted to the Nanyang Technological
University in partial fulfilment of the requirement for the
degree of Doctor of Philosophy

2018

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Chapter 3.1 is published as Yu Zhou, Ziyu Wang, Abdullah Rasmita, Sejeong Kim, Amanuel Berhane, Zoltan Bodrog, Giorgio Adamo, Adam Gali, Igor Aharonovich, Wei-bo Gao Room-temperature solid state quantum emitters in the telecom range *Science Advances* **4** 3, eaar3580 (2018).

The contributions of the co-authors are as follows:

- Prof. I.A., Prof. W.-B.G. supervised the project.
- I performed all the experiment at the School of Mathematical and Physical Science with Ziyu Wang.
- A.R., and S.K. and me analyses the data from the experiment.
- Z.B. and A.G. did the model simulation work.
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- Prof. Jianming Cai, Prof. W.-B.G. supervised the project.
- I performed all the experiment at the School of Mathematical and Physical Science with Junfeng Wang and Xiaoming Zhang.
- Junfeng Wang, Xiaoming Zhang. and Ke Li and me analyses the experiment data.

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Abstract

Solid-state single-photon sources are the central blocks for many scalable quantum applications including quantum computing, quantum key distributions, and quantum simulations. Over the years, diverse quantum emitters have been studied. This thesis endeavored investigations into single-photon sources in diamond, silicon carbide and gallium nitride in three directions:

Firstly, identification of the single-photon sources that have never been reported before and fabrication technique for scalable single germanium vacancy centers in diamond. Telecom wavelength single photon sources have been found in both gallium nitride and silicon carbide. More importantly, those emitters work even at room temperature and are promising for quantum key distribution applications in the future.

Secondly, the spin properties study of the single silicon vacancy in diamond: the silicon vacancy optical transition has been coherently controlled in the work. The optical Rabi oscillations in the time domain have been observed. The ultrafast spin control of the silicon vacancy has been also achieved.

Thirdly, quantum applications of the divacancy spins in silicon carbide: nano-scale thermometry based on the quantum control of the divacancy spin in silicon carbide has been introduced. Especially, we found a self-protected effect that the transverse strain can protect the spin from the local magnetic field and enables longer coherence time.

Finally, some possible directions have been discussed.

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First author:

1. **Yu Zhou**, Ziyu Wang, Abdullah Rasmita, Sejeong Kim, Amanuel Berhane, Zoltan Bodrog, Giorgio Adamo, Adam Gali, Igor Aharonovich, Wei-bo Gao Room-temperature solid state quantum emitters in the telecom range *Science Advances* **4** 3, eaar3580 (2018).
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Chapter 1 Introduction

1.1 Solid-state single photon sources (SPS)

Single photon sources are the light source whose emission exhibits quantum nature that the time interval between two successive photons is larger than a minimum value. It is distinct with the laser emission whose photon statistical distributions follows a Poissonian distribution and thermal light source whose photons are tending to be bunched together [1]. Such single photon source has been proposed or demonstrated in various of quantum applications like quantum simulation (such as boson sampling [2]), quantum metrology [3], quantum computation [4-5] and quantum communication [6].

SPS was first demonstrated in the optical transition in sodium atoms [7]. Now the technique to produce single photon based on the atom system is already advanced [8]. However, the whole system is demanding in terms of high vacuum and a complex control scheme. Other methods like spontaneous parametric down-conversion [9] and four-wave mixing [10] have also been employed to generate single photons, but the purity of single photons is a problem.

Decades of efforts have been put into searching a suitable solid-state system that host superior SPS [11-13] with excellent optical properties. They exist in the solid-state materials like diamond, silicon carbide, gallium nitride or can be self-fabricated. The tradeoff is that the mesoscopic environment of the solid will induce both homogeneous linewidth broadening and inhomogeneous distributions which degrades the quality of the SPS [14]. However, those side effects can be further mitigated by various techniques

[14].

1.1.1 Defects in diamond

Diamond is not only for jewelry, researchers found that it is also a host for many kinds of single photon emitters [15]. Lattice is not always perfect, it is common that in some lattice, carbon atoms are missing or replaced by another element. For example, if many carbon atoms are replaced by nitrogen, the color of the diamond will be yellow. The imperfection of the lattice results in these defects. Several different types of defects have been investigated, such as nitrogen vacancy center (NV center), silicon vacancy center [16], germanium vacancy center [17], Sn vacancy center [18] etc.

1.1.1.1 Nitrogen vacancy center in diamond

NV center in diamond is the first well-studied solid-state quantum emitter. As shown in Figure 1-1 [19], the atomic structure of the NV center. It consists of one missing carbon lattice site (vacancy) bounds to another substitute nitrogen atom. Figure b is the photoluminescence measurement of the NV center. Zero phonon lines of two different charge states of NV center: NV^0 and NV^- are marked with yellow and red arrows respectively. Figure c is the energy level of the NV center. The green arrow represents the off-resonant excitation. Here beside ground state 3A and excited state 3E , another intermediate dark state 1A is introduced to explain the spin polarization mechanism. Without the magnetic field, NV center is spin triplet system with $m_s = 0$ and $m_s = \pm 1$ (degenerated). When $m_s = \pm 1$, besides the normal decay channel between ground and excited state, it has a higher possibility to go through the non-radiative process to the $m_s = \pm 1$ the thicker grey dashed arrow shows. On the contrary, when $m_s = 0$, it is less likely to go to the intermediate dark state 1A . This process will result in

two consequence: 1. The emission will be stronger when $m_s = 0$ compare to $m_s = \pm 1$. 2. When the system is continuously excited with a green laser, all the population in $m_s = \pm 1$ will be transferred to $m_s = 0$ and finally, $m_s = 0$ will dominate and spin initialization can be achieved with green laser illumination. The zero-field splitting between $m_s = 0$ and $m_s = \pm 1$ is around 2.87GHz, if the microwave field with the exact frequency is added, the spin manipulation can be realized. This is the fundamental reason of how Optically detected Magnetic resonance (ODMR) works as shown in Figure 1-1d. When the magnetic field is zero and if the microwave field is not on resonance with the energy gap between $m_s = 0$ and $m_s = \pm 1$, the spin will not be flipped, and the fluorescence will not change. Once it is on resonance, $m_s = 0$ is driven to $m_s = \pm 1$ which will result in a fluorescence decay as the grey curve shows. If a static magnetic field is added, the $m_s = \pm 1$ will split as Zeeman splitting happens with a slope of 2.8MHz/G.

Figure 1-1 NV centers in diamond. (a) The molecular structure of nitrogen-vacancy center in diamond. It consists of a vacancy (white ball) and a substituted nitrogen atom. (b) Photoluminescence

measurement of the NV center under off-resonant excitation. The zero-phonon line of NV^0 and NV^- are marked with yellow and red arrows respectively. (c) The energy level of the luminescence process when excited with off-resonant laser field (green). (d) Optically detected Magnetic resonance (ODMR) measure of a single nitrogen vacancy center, when different B field was added, the Zeeman splitting will appear [19].

1.1.1.2 Silicon vacancy center in diamond

Although NV has attracted plenty of interest, researchers are still looking for a better candidate for only 4% photons lie in the zero-phonon line region which limits its quantum applications and coupling to photonic structures such as nanocavities and waveguides. On the contrary, more than 80% of photons of silicon vacancy lies in the zero-phonon line and it has many other superior optical properties such as lifetime limited line width, the feasibility of optical and microwave control of the spin states and etc [20].

Lifetime limited linewidth

The linewidth of a solid-state emitter is an important parameter. Narrower linewidth which ensures longer coherence of the photon is preferred. However, the microscopic environment of the defect will induce many broadening mechanisms. There are two major ones: homogenous broadening and inhomogeneous broadening. Homogenous broadening means all defects will be affected by this broadening like lifetime broadening which can be explained by the uncertainty principle. Also, the pure dephasing of the ground and excited state will also be contributed to the broadening. Inhomogeneous broadening is more localized and varies from defect to defect. Such as localized Stark shift or strain. In silicon vacancy, lifetime limited linewidth has been reported in silicon vacancy in diamond [21].

Figure 1-2 Transformed limited linewidth in silicon vacancy in diamond (a) Photoluminescence measurement of a single silicon vacancy center in diamond at both 4K and 50K. (b) The energy level structure of the silicon vacancy with ground and excited state splitting 46.7 GHz and 258.1GHz respectively. The ABCD transitions in (a) are marked as green and pink arrows respectively. (c) Resonant excitation of ABCD transitions, the raw dots are the raw data and the solid lines are the Lorentz fitting. (d) The lifetime measurement of the single SiV at both 4K and 50K [21].

As shown in Figure 1-2, silicon vacancy center in diamond has characteristic ABCD four transitions energy level structure. The ground and the excited state will split due to the spin-orbit coupling. The ground and excited splittings are 46.7 GHz and 258.1 GHz respectively. The linewidths are measured with resonant excitation and for the D transition, the fitted linewidth is about 119 MHz which is close to the lifetime limited value $1/2\pi T_1$ which is roughly 92.5 MHz.

The lifetime broadening mechanism dominates in silicon vacancy which can be proved by the observed lifetime limited linewidth. Others like pure dephasing and strain contribute little to the broadening which ensures the long coherence of these photons and can be more efficient coupling to the photonic structures like nanocavities and waveguides.

Optical and microwave coherent control

As mentioned above, NV attracts the most attention because its spins can be initialized and read out optically and can be controlled with the microwave field. Both optical control and microwave control of the silicon vacancy has been achieved.

Figure 1-3 Energy structures a. The energy level of a single silicon vacancy center in diamond. The ground and excited state split 48GHz and 256GHz respectively because of spin-orbit coupling. ABCD are four characteristic transitions. b. The resonant excitation spectrum of a single SiV in diamond [22].

The first realization of spin control in silicon vacancy in diamond is achieved by using ultrafast laser pulse like picosecond pulse [22]. If one of the ABCD, as shown in Figure 1-3, is excited with picosecond laser, by varying the pulse area of the ultrafast laser field, the population of the excited state will undergo oscillations so that one photon Rabi oscillation will be achieved. As displayed in Figure 1-4, the fluorescence oscillates with the square root of the picosecond laser which is on resonant with transition C and D respectively.

Figure 1-4 Optical one-photon Rabi oscillations a. Fluorescence with respect to the square root of the power of the picosecond laser which is resonant with transition C and transition B respectively [22].

Besides ultrafast optical pulses, microwave field is another method to control the spin [23]. As shown in Figure 1-5, the spin was first initialized with a 737nm pulse and read out by another 737 nm pulse. Between these two pulses, a microwave field was added. Once the microwave is on resonance with the energy level gap as shown with green and yellow circle in Figure 1-5b, the microwave will transfer the population of the spin up and spin down states induced by a static magnetic field and will result in peaks in the ODMR spectrum in Figure 1-5c. The energy structure can be also verified by applying a magnetic field. Due to Zeeman splitting, the ODMR spectrum will also split as Figure 1-5d shows.

Figure 1-5 Optically detected magnetic resonance (ODMR) a. The pulse scheme of the ODMR detection, a 737nm pulse was first to initialize the spin and the second was to read out the spin states. Between these two pulses, a microwave pulse was added. b. The energy level of one ground states (are split by magnetic fields (blue spin down and red spin up). The further fine structure was induced by the coupled nuclear spin. c. When the microwave is on resonant with the energy splitting in b, the population transfer will happen which induce a peak on the ODMR spectrum. d. When the magnetic field is added, the ODMR frequency will shift because of the Zeeman splitting [23].

Recently, similar as silicon vacancy, the Ge, Sn, and Pb center have been discovered. Those interstitial atoms are in the same column and those emitters share similar optical properties like ABCD structures, high Debye-Waller factor etc. However, the energy splitting between ground and excited state is different. The larger splitting is preferred because of it may block the phonon mediate effect thus ensure longer coherence time.

1.1.2 Defects in silicon carbide

1.1.2.1 Silicon vacancy in silicon carbide

Silicon carbide is a well-known semiconductor material which is widely used in modern industry. Unlike diamond, silicon carbide has mature fabrication technique and will benefit future quantum qubits on-chip integration.

Silicon vacancy is one of the room temperature addressable spins in silicon carbide.

The coherent control of single silicon vacancy center was first reported in [24] as shown in Figure 1-6. Figure 1-6a is the photoluminescence measurement of a single SiV, the spectrum is from 750nm to 1050nm which is in the near infrared region. To confirm it is single SiV, second-order autocorrelation measurement was done and $g^2(0)$ was well below 0.5. To enhance the collection efficiency because of the high insertion loss due to the high refraction index, a solid immersion lens (SIL) was made. A factor of 3 enhancement has been observed. Figure 1-6d shows that the emission was stable without any blinking or bleaching.

Figure 1-6 Single SiV in silicon carbide at room-temperature a. The PL spectrum of single SiV in silicon carbide. b. Second-order autocorrelation measurement to confirm the single photon nature. c. the comparison of the saturation curve of single SiV when SIL is implemented or not. d. The fluorescence counts with respect to the time [24].

1.1.2.2 Divacancy

Figure 1-7 Divacancy spins atomic structure in silicon carbide Due to the crystal structure, the individual lattice has hexagonal (h) or quasi-cubic(k) type. So, the divacancy will be in the different forms like kk, kh, hk and hh type [25].

Divacancy is the defect whose both two adjacent atoms are missing in the crystal lattice. For Example, in the silicon carbide lattice structure as Figure1-7, neighboring silicon and carbon atoms are missing forming divacancy. Moreover, because of the crystal structure, there are two different lattices namely hexagonal (h) and quasi-cubic (k) position. Thus, divacancy has different types due to the missing atom at a different position, such as kk, kh, hk and hh.

Figure 1-8 Divacancy spins in silicon carbide a. The PL spectrum of divacancy ensembles in

silicon carbide. b. Zero phonon lines position of different divacancy PL1-PL7 c-h. Each divacancy's (PL1-PL7) zero phonon line was filtered out to measure ODMR spectrum individually [25].

Divacancy spin Coherent control of divacancy spins in silicon carbide was first demonstrated in 2011 in [25]. As displayed in Figure 1-7. The temperature dependence of the PL spectrum was measured in Figure 1-7a, each zero-phonon line corresponding to a different type of divacancy. To do the ODMR individually, one must isolate those spins. One way is to filter out each zero-phonon line by filters or gratings and perform ODMR of PL1-PL7 respectively as Figure 1-8 c-h.

Figure 1-9 Coherent control of divacancy in silicon carbide a. Rabi oscillations of divacancy spin in silicon carbide. b. Rabi frequency with respect to the power increase. A linear relationship is clear [25].

The coherent control of divacancy has been demonstrated in Figure 1-9, the microwave frequency is 1.3526 GHz and the Rabi oscillation is clear. To confirm the coherent control of the spins, the frequency of the Rabi oscillation is linear with the square root of the power microwave applied.

1.1.3 Other emerging solid-state SPSs

SPSs has been reported in other systems like two-dimensional materials [26-27], hBN flakes [28], rare earth ions in crystals [29] and carbon nanotubes (CNTs) [30].

There are several criteria to judge if the SPS is superior in optical properties: First, the fluorescence intensity. To achieve real applications like quantum key distributions (QKDs), the count rate should reach a high level. Second, the homogeneous linewidth at 4K should be narrow. This ensures the long coherence of the photon stream and the generation of Indistinguishable photons. Third, if we have the method to spatial and deterministic generate them, this is crucial to the large scalable integration to the real quantum applications. Finally, if the SPS is easy to be coupled with photonic structures that makes their optical properties more attractive. Till now, there is no one SPS that is perfect in all criteria so that much efforts have been still putting into searching a better SPS.

1.2 Solid-state spins and its quantum applications

1.2.1 Quantum key distribution

Attenuated laser pulses were used in most QKDs demonstrations for convenience. However, the average number of photons in each pulse needs to be controlled and the sifted key is required to be shrunk because of the statistical effects from the finite key size[31]. Moreover, it has some security problems that the source attack can happen once the hacker gets the phase information [32-33]. So the true single photon source is still preferable in QKDs for simplicity and security problems.

QKD over 120km based on SPS in solid has been demonstrated in 2015 in [34]. As Figure 1-10 displays, the QKD experimental scheme. The source is telecom-band InAs/InP quantum dot which operates at cryogenic temperature. The repetition rate can reach up to 62.5M. They used a time-bin based encoding BB84 system [35-37].

Figure 1-10 QKD experiment set up [38]

1.2.2 Quantum network and computing

Figure 1-11 Quantum network [39]

Quantum network was first proposed in 2008 in [39]. In the Figure 1-11, the quantum network is composed of two essential elements, first is the quantum node and second is the quantum channel. The quantum nodes are used to store and process the information. Between those quantum nodes, there are quantum channels which transfer information between quantum nodes. For the quantum channel, the photon is the best candidate for they are traveling fast, and photon streams can carry much encoding information based on their colors, polarizations and etc. And for quantum nodes, solid-state quantum spins are one of the candidates. As I mentioned above, spin initialization, manipulation and read out has been demonstrated already in some quantum spin system.

So those spins are suitable to act as a stationary quantum information processing node, however, how those spins can talk with each other or realization of multi-entanglement is still a challenging thing.

Figure 1-12 Quantum entanglement between two NV spins separated by 1.3km. In the picture, station A and station B are where the individual spin locates and their emission is guided in the optical fibers which is marked with dashed red lines [40].

The generation of robust entanglement between two NV center spins that separated 1.3km [40] as shown in Figure 1-12. The Bell inequality was tested again in this experiment without any additional assumptions.

1.2.3 Quantum sensing

Spins in the solid are not only for quantum register as mentioned above. They can be very precise sensors.

Temperature sensing

High spatial resolution and high accuracy thermometry are always demanding in biological applications or in the industry. Many efforts have been put in this direction. The localized temperature sensing has been already demonstrated based on many systems like Raman technique, scanning probe microscopy, fluorescent nanoparticles

and etc. However, many of them suffer from low sensitivity or sophisticated experimental schemes. Solid state emitters like NV center in diamond has been also used to be a temperature sensor. The zero-field splitting (D value) of the NV center is around 2.87 GHz at room temperature. However, this value is not fixed, and researchers found it is linear with temperature from 280K to 330K [41], so if one can precisely measure the D value, the temperature of the system can be sensed by the former calibration. Moreover, the spatial resolution of this technique can reach to hundreds of nanometers which is the diffraction limit of the confocal microscopy set up. The nanometer scale thermometry in a living cell was demonstrated in 2013 in [42]. As shown in Figure 1-13, many nanodiamonds containing NV centers are swallowed by the cell, and a gold nanoparticle is inside the cell for the purpose of heating. In the experiment, thermometry has been performed in A-F point, in this order distance from the nanoparticle is increasing. The temperature shows a decreasing trend as expected in Figure 1-13d. The temperature was measured by precisely fit the ODMR spectrum in the figure 1-13a, four marked red dots are used in the fitting.

Figure 1-13 Left: Nanometer scale thermometry in a living cell. a, ODMR measurement of a single NV center in nano diamond. The four red dots are picked up to fit the center frequency precisely. b, the confocal scan map of the living cell, the hit point is gold nanoparticle for heating with the laser beam. A-F are the points where thermometry performed. c, when the heat power increases the sensed

temperature also rises linearly. d. Temperature sensing for A-F, as the distance from the point to the nanoparticle increase, the temperature decreased as expected [42]. **Right: Thermal Echo sequence.** a, The thermal echo sequence that used in the thermometry. b, the population of the spin under the thermal echo pulse, the oscillation is due to the detuning from the ODMR on resonance value c. the period of the oscillation changed because of the temperature increase 0.1K, the thermometry was realized based on this period change of the thermal echo [43].

Besides by measuring the exact ODMR frequency of the system to realize the thermometry. There is another way that developing a brand new control sequence of the spin that can reflect the ODMR resonance [43]. By detecting the oscillation period of the thermal echo which is closely related to the temperature. As shown in Figure 1-13, A is the thermal pulse. A $\pi/4$ pulse was added on the system and then wait for the system to evolve a period of time τ . Then a π pulse was added and let the system evolve another τ . The microwave frequency was intentionally detuned from the exact resonance for the oscillation like B occurs. If the temperature was increased 0.1K, the ODMR resonance will be changed which will also induce the change of the oscillation period as shown in Figure C.

Magnetic sensing

High spatial resolution magnetic imaging is also a demanding technique in many areas like biology or material sciences. The NV is the emerging sensor considering its nanoscale size and high sensitivity to the outside magnetic field. The magnetism is not complicated, under B field, the ODMR spectrum will split at a slope of 2.87MHz/G, by precisely measuring the ODMR resonant center frequency, the B field can be extracted. The diamond cantilever which contains single NVs will be attached to an AFM tip and will be very close to the sample (hard disk or some magnetic material). The whole sample was cooled into 6K and the excitation was done above the sample and AFM tip.

The magnetic image of the hard disk is shown in Figure 1-14 b and c [44].

Figure 1-14 Cryogenic NV scanning probe magnetometry. a, The scanning probes is the diamond cantilever attached in the AFM tip, in each cantilever, there is a single NV center and when the manometry is performing, only one NV center will be chosen. b, The bit structure of the hard disk. c, the vortices in BaFe_2 [44].

1.3 Motivation and Objectives of this Thesis

The motivation of the thesis is to investigate several questions: (1) How to deterministically generate a large number of SPSs in a scalable manner. (2) Is there any more superior SPSs than those who have been reported? (3) How to realize the spin control in some SPSs that never been tried before? (4) What kind of real quantum applications that SPSs have?

The objectives of the thesis are clear: to summarize the reported SPSs and introduce my work about new founded SPSs that emits at the telecom range which is desirable in the area of quantum communication. Moreover, the scalable production of SPSs is still a challenging thing, direct writing of single Ge center in diamond has been introduced

in this thesis. What's more, for some SPSs, the energy level and the physics behind it is still unclear and worth further investigation, our work about coherent control of silicon vacancy in diamond will be introduced. Finally, to figure out more quantum application of solid-state spins, like our work the nanoscale thermometry with divacancy spins in silicon carbide will be introduced.

1.4 Organization of This Thesis

In Chapter 1, I will give an introduction to single photon sources. I will introduce the main type of SPS that have been reported, such as defects in diamond, defects in silicon carbide and other SPSs. Moreover, I will introduce some real quantum applications of the SPSs in the field of (1) quantum key distribution. (2) quantum computation. (3) quantum sensing.

In Chapter 2, I will describe the equipment for my study, including the cryogenic setup and home-made confocal microscopy system I built. Also, the microwave set up which is used to control the spin will be introduced.

In Chapter 3, I will present my Ph.D. work on the identification and fabrication of solid-state SPEs. Three main works will be introduced. The telecom wavelength single-photon emission has been found both in gallium nitride and silicon carbide. And scalable fabrication of single germanium vacancy center will also be introduced.

In Chapter 4, I will present my Ph.D. study on an example of spin control of SPE: coherent control a strongly driven optical transition in silicon vacancy in diamond. The optical Rabi oscillations and Mollow triplet have been observed. Theoretical study of the three-energy level system in silicon vacancy agrees well with our experimental

observations.

In Chapter 5, I will present my Ph.D. work about an example of the real quantum application of spins in the solids: self-protected nanoscale thermometry based on divacancy spins in silicon carbide.

Finally, I will draw some conclusions and discuss the future possible directions in Chapter 6.

Chapter 2 Instrumentation and principles

2.1 The confocal microscopy system for single photon emission detection

Figure 2-1: Left :Home-built confocal microscopy system [45]. Right: Real image of confocal microscopy system.

The home built confocal microscopy system is the key set up to measure SPE. The excitation and collection arms are confocal which means the excitation beam and the collection arm is focused on the same area on the sample. The emission which has been excited by the focused laser beam will be collected by the collection arm. As displayed in Figure 2-1, the procedure is as follow: the excitation beam from the diode laser is guided to the entrance of the excitation arm and become a collimated beam through a selected lens. Then the beam will be purified and reflected at the dichroic mirror. The dichroic ensures that the shorter wavelength beam is reflected (the excitation) and the longer wavelength beam is passed (the collection). The excitation laser will be focused onto the sample in a few micrometer level sizes by passing through an objective and excite the SPE. Then the emission of the SPE will follow the sample path and then pass the dichroic mirror again. Finally collected by the optical fiber. The optical fiber will

be connected to either spectrometer for PL measurement or APD for counting purpose. For the photons in the visible range, Single Photon Counting Module (SPCM-AQRH) whose photon detection efficiency greater than 70% at 700nm. The dead time is around 30ns based on the specification. For the photons in the infrared range, a superconducting detector (Scontel) is used. The Quantum Efficiency is more than 85 % from 0.7um to 1.3um. The timing jitter is less than 45 ps and dark count is 10cps. The sample was mounted on the xyz piezo stage with nanometer spatial resolution. So, an XY scan map can be got if the stage scan in xy plane and record every pixel's photon counts as shown in Figure 2-2.

Figure 2-2: Typical confocal scan map which is the contour graph of the photon counts in every pixel.

The green or red dots are the pixels where collected photons are more than those blue parts, the green and red dots in the graph is where SPE should locate.

2.2 Spin initialization readout and manipulation

Photoluminescence is measured by the spectrometer which has optical grating in it to spread out the different color of light and measured by CCD camera as shown in Figure 2-3 left. The spectrometer is equipped with two liquid nitrogen cooled camera (PYL100BR Princeton Instrument for visible measurement and PyloN-IR Princeton Instrument). In the spectrometer, there are three gratings installed (300 I/mm, 750I/mm and 1200 I/mm) with different spectral resolution. Usually the photoluminescence consists of two parts, zero-phonon line and phonon sideband parts as displayed in Figure 2-3. Due to the thermal effect, most of the zero-phonon line will disappear or broaden in the cryotemperature, so zero phonon line at cryogenic temperature is regarded as one of the signatures of an emitter. For example, the right figure of Figure 2-3, V1(860nm) and V2(917nm) are regarded as the signature of silicon vacancies in 4H silicon carbide.

Figure 2-3: Left: Spectrometer. Right: Photoluminescent measurement of silicon vacancy in silicon carbide in 5K. [46]

2.3 Second-order autocorrelation measurement

To prove if the emitter is single photon emission, a second-order autocorrelation

function $g^2(\tau)$ is usually measured.

$$g^2(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2} = \frac{\langle E^*(t)E^*(t+\tau)E(t+\tau)E(t) \rangle}{\langle E^*(t)E(t) \rangle^2}$$

The $\langle \rangle$ is the time average value. $I(t)$ represents the intensity at time t . And E is the electric field and its conjugate is E^* .

Figure 2-4: Different types of photon streams (antibunched, coherent, bunched)

[47].

Figure 2-4 is the simple illustration figure of three different photon streams [47]. If the light is perfect coherent light, the time intervals between two adjacent photons follow Poissonian distribution and it is totally random. So the $g^2(0) = 1$ in the coherent light case. If photons are likely to bunched together, if the detector detected one photon, then the possibility of detecting another photon in short time is larger than in a long time. It will result in $g^2(0) > 1$, also called bunching effect. If all photon has an interval between photons, these photons come out one by one. Then if one detector detected the photon, the possibility that another photon will come in a short time (less than the time gap) is zero. One would find that $g^2(0) < 1$, called antibunching effect. For a perfect

single-photon emitter, $g^2(0) = 0$.

Figure 2-5: A. Simple diagram for second-order autocorrelation measurement. B. An example of second-order autocorrelation measurement for SPE found in gallium nitride[48].

Second order correlation measurement can be measured by Hanbury-Brown-Twiss set up as shown in Figure 2-5A. A 50:50 beam splitter was used to split the incoming photon stream and to two photon counters. One APD is used as a start signal, and the other one is stop signal. The time difference τ between start and stop will be analyzed in the histogram like in B. The blue dots (raw data) without any background correction is fitted with a three-energy level system as the red solid line shows, we got the $g^2(0) = 0.05 \pm 0.02$, which shows that the emission comes from a single photon emitter whose photon statistics has a clear anti-bunching effect.

2.4 Spin initialization readout and manipulation

To build a quantum computer, David DiVincenzo listed the following requirements in

2000[49]:

1. A scalable physical system with well-characterized qubits.
2. The ability to initialize the state of the qubits to simple fiducial state.
3. A "universal" set of quantum gates.
4. Long relevant decoherence times, much longer than the gate-operation time.
5. A qubit-specific measurement capability.

Next, I will show that the spin initialization, manipulation and read-out have all been achieved for some solid-state defects. However, it is still a big challenge to realize multi-qubit gates in a scalable manner.

2.3.1 Spin initialization

Figure 2-6: The energy level diagram for the NV center[15].

The electronic structure of NV center is as displayed in Figure 2-4. It involves ground state 3A_2 , excited state 3E and intermediated dark states 1A_1 and 1E . The all the transition has been marked with the dash (weak) and solid (strong) arrows. The ground state 3A_2 and excited state 3E are spin triplet states who has $m_s = 0$ and ± 1 . $m_s = \pm 1$ degenerate.

If the system is under green (532nm) laser excitation, it prefers to stay in $m_s = 0$ rather

than $m_s = \pm 1$. So after some cycles, all the population will be initialized to $m_s = 0$ very efficiently.

2.3.2 Spin readout

As shown in Figure 2-6, if the electronic state is in the $m_s = 0$ state, the radiative cycle will result in stronger fluorescence than $m_s = 1$ state. The difference is roughly 30% [15]. Thus, by comparing if the fluorescence is bright ($m_s = 0$) or dark ($m_s = 1$) the spin can be read out.

Figure 2-7 Single shot read out of the nuclear spin in diamond. A. The simple scheme for reading the nuclear spin. B. The readout of a nuclear spin reveals a quantum jump of the nuclear spin [50].

Single shot spin readout has been also achieved in the NV center system. As displayed in Figure 2-7, A CNOT gate has been implemented. It is realized by nuclear spin selective ($m_n = -1$) microwave flip the electron spin to its $m_s = -1$ state. If the nuclear spin is indeed -1, and the flip of the electron spin will result in a fluorescence drop. Otherwise if the nuclear spin $m_n = 0$ or $+1$, the fluorescence level will remain. So, the telegraph-like signal in Figure 2-7B is the indication of whether the nuclear spin is in -

1 (high level) or other states. That is how single shot readout of nuclear spin has been achieved.

2.3.3 Spin manipulation

ODMR (Optically Detected Magnetic Resonance) is a technique that used to study the electron spin by the optical method. A microwave field will be fed to the vicinity of the defects. If the microwave frequency is on resonance with the energy gap, the spins states will be transferred which will be monitored by fluorescence change.

Figure 2-8 ODMR spectrum of NV center[15].

Take NV center as example, figure 2-8 shows the typical ODMR spectrum of NV center in diamond. When the microwave is on resonance with its energy splitting between $m_s = \pm 1$ and $m_s = 0$, the fluorescence will decrease which is a deep in the ODMR spectrum. (1.42 GHz for excited states splitting and 2.87 GHz for ground state splitting).

Figure 2-9 Pulse scheme for Rabi and Ramsey measurement in PL5 defects in silicon carbide.[51]

Two basic qubit operations are Rabi oscillations and Ramsey fringes. The scheme is as shown in Figure 2-9, a 2μs laser pulse is used to initialize the spin and the second 2μs laser pulse will be used to read out the spin state. Between the two optical pulse, microwave pulses either a pulse vary the duration for Rabi or two $\pi/2$ pulses for Ramsey.

Chapter 3 Identification and fabrication of solid state SPSs

As I introduced in chapter 1, many types of SPS have been reported and there is no single SPS that is excellent in every aspect, so searching for a good SPS, especially for some special property that can be used in a specific quantum application is still an important topic. In this chapter, I will introduce our work on new observations of two telecom wavelength SPSs both in gallium nitride and silicon carbide. Moreover, the scalable and direct writing of a single germanium vacancy center will also be covered.

3.1 Room-temperature solid-state quantum emitters in the telecom range

Single photon sources (SPSs) play an important role in many quantum applications like quantum networks and QKD protocols. In which the photons can be regarded as flying qubits which bring information with them. And people prefer those photons whose energy lies in the telecom range because of the minimum transmission loss in the optical fibers. Although a lot of efforts have been put into finding an SPS which operates at room temperature and at the same time emits at the telecom wavelength. In this part, I will present our findings of room temperature SPSs in gallium nitride- a well-known semiconductor material. The emitters we found has high single photon purity and a very high brightness of ~ 1.5 MHz. These SPSs may help the development of on-chip quantum technology applications.

3.1.1 Motivation to find a suitable room temperature and telecom wavelength SPS

In the last decade, SPSs have been demonstrated as a key element for many quantum applications [52-58], including quantum simulation [59-61], quantum metrology [62]

and quantum computing in which photons can be used as “flying qubits” which carries information [63]. In the case of quantum computation in the linear optics region, single photons interact with each other with different projective measurements [63]. Moreover, single photons from SPS can be used as inputs for different interferometers and form complete photonics circuits in boson sampling experiments [2]. Single photon sources are the key element in the QKD schemes. But because of the lack of a practical ultra-bright SPS, weak coherent pulses from attenuated lasers are still widely used in current QKD [64]. A true SPE would still be a preferable because of the security problem.

In all the applications mentioned above, the telecom wavelength photons [65] is preferred because of the lower transmission loss in the optical fibers than the other wavelengths. Semiconductor quantum dots might be an option, for example, InAs/InP QDs emits at the telecom wavelength and have been used to realize 120 km QKD [34]. However, the most disadvantages of them is the requirement of cryogenic temperatures, which is not convenient. In another approach, the non-telecom wavelength can be under down conversion scheme. However, the conversion efficiency of the non-linear process is a problem. Finally, very recently, SPSs in the infrared spectral range has been found in carbon nanotubes (CNTs), but the emitters suffer from blinking and low count rate [30].

In this chapter, I will present a room-temperature SPSs in the bulk GaN at the telecom range. The single photon purity can reach 0.05. What’s more, they are extremely bright, the count rate reaches up to 10^6 counts per second. Many kinds of defects in the UV or visible range have been studied extensively before [66-67]. However, no defects at the telecom wavelength has been discovered. Moreover,

considering that the production and fabrication of the gallium nitride nanostructures are advanced and thus the discovery of telecom SPSs can be instant to the on-chip integration with the help of the up to date high technologies.

3.1.2 Gallium nitride SPSs in the telecom wavelength

Figure 3-1. Infrared SPSs in GaN. (A) Atom structure of the bulk gallium nitride and the real image of the gallium nitride wafer. (B) The confocal scan map the gallium nitride where an SPS is at the center. (C) 6 SPSs' PL measurement at both low and high temperatures indicating that the PL has a distribution which ranges from 1085 nm to 1340 nm. (D) Second-order correlation measurement for the SPS 1 with 950 nm continuous wave laser excitation. The blue dots are the raw data and is fitted with a three-energy level system as the red solid line shows, we got the $g^2(0) = 0.05 \pm 0.02$. (E) Second-order correlation measurement of the SPS 1 with an 80Mhz repetition rate pulsed excitation. From integration of the blue area, we get $g^2(0) = 0.14 \pm 0.01$. This measurement is done in the room temperature.

Figure 3-1A shows the gallium nitride atomic structure and a real image of our GaN wafer which has a (Mg)-doped GaN layer and undoped GaN layer grown on sapphire, the thickness of both is 2μm. Firstly, we measured the PL measurement of the GaN SPSs with our confocal microscopy system, the excitation was done by a 950nm diode laser and the emission was collected with optical fibers to superconducting

detector. A Hanbury Brown-Twiss (HBT) was built for the photon correlation measurement or a spectrometer for PL measurement. As displayed in Figure 3-1B, SPSs are randomly distributed and at the center, the single photon emitter is marked as SPS which emits 350 kcounts per second at the room temperature. We measured different SPS's PL and found that each emitter has its own zero-phonon line and the ZPL energy ranges from 1085 nm to 1340 nm as Figure 3-1C shows. At room temperature, the full width at half maximum (FWHM) of the emitters' linewidth is also different from emitter to emitter, the minimum one is 3 nm (ZPL at 1120nm), and the maximum one is 50nm (ZPL at 1285nm). When the sample was cooling down to 4K, the FWHM of the measured SPSs reduces to a few nm, but it is still quite broad, it is probably due to the coupling to the lattice.

We then measure the second order correlation function of the SPSs we found with both CW and pulsed laser excitation. As the Figure 3-1D shows, the second-order correlation function $g^{(2)}(\tau)$ has been recorded for SPS 1 with CW laser excitation. The dip at the position of zero delay time indicates that the emission is non-classical and the source is indeed a single photon emission. The blue raw data is fitted with a three-energy level model with equation

$$g^2(\tau) = 1 - \alpha * \exp(-|\tau|/\tau_1) + \beta * \exp(-|\tau|/\tau_2)$$

in which τ_1 is the excited state lifetime and τ_2 is the metastable state lifetimes, respectively, α and β are the fitting parameters. The obtained $g^2(0)$ is 0.05 ± 0.02 , suggesting that the single photon purity is high. And we want to stress out that the deviation from exact zero is because of the emission from other emitters is also coupled to the optical fiber. We have also measured the second order correlation function in

the pulsed laser excitation case. As displayed in Figure 3-1E, the SPS can be triggered efficiently. To obtain the $g^{(2)}(\tau)$ in a pulsed regime, the picosecond pulse with a repetition rate of 80MHz was used to excite the SPS and the obtained $g^2(0) = 0.14 \pm 0.01$ by the integration of all the blue part. It is also well below 0.5 suggesting that it is single photon emitter. The value of the $g^2(0)$ is higher than the CW case because of the higher backgrounds in the pulsed excitation regime.

Figure 3-2. Optical characteristics of SPS 1. (A) Saturation measurement of SPS 1, the black dots are the raw data and the red curve is the fitting. From the fitting with the function $I(P) = I_{\infty} \times P / (P + P_s)$, we got saturation power is $P_s = 2.32$ mW, and saturation counts $I_{\infty} = 0.69$ M counts/s. (B) The SPS was excited at three different power (0.1 mW, 0.8 mW and 1.5 mW), the total acquisition time is 2 minutes and the time is 100 ms. (C) Lifetime measurement of SPS with a pulsed laser, the black dots are the raw data and a single exponential decay was used to fit the red data. We got a lifetime $\tau_1 = 736 \pm 4$ ps. The blue curve is the measured superconducting detector's response function. (D) A schematic diagram of a three-level system to explain the observed bunching effect. (E) The excitation and emission polarization pattern of SPS1.

To characterize the brightness of the SPSs, a saturation curve is measured for the SPS 1 as displayed in Figure 3-2A. The data is fitted well with $I(P) = I_{\infty} \times P / (P + P_s)$, in which I is the emission rate when the excitation power is P . Saturation count I_{∞} and saturation power P_s are the free parameters. From the fitting, we get the saturation

power P_s was about 2.32 mW, and moreover, the saturation counts is around 0.69M counts per second. Such a high-count rate is comparable to the most bright emitter found in the solids. I will discuss the extraction efficiency in the later section.

We then measured the photostability of SPS 1 as displayed in Figure 3-2B with 0.1s time bins and 2 minutes duration at different powers 0.1 mW, 0.5 mW and 1.8 mW with different colors. No obvious blinking or bleaching has been observed in the measurement which proves that it is single emission. The lifetime measurement of SPS 1 is presented in Figure3-2C. From a single exponential fitting, we obtained the lifetime 736 ± 4 ps , which is comparable to the value obtained from the $g^{(2)}(\tau)$ fitting (776 ± 39 ps). A three-level model (Figure 3-2D) is need to explain the observed bunching behavior. Both excitation and emission's polarization are shown in Figure 3-2E fitted with $\cos^2(\theta)$. The polarization visibility which is calculated by $\eta = (I_{max} - I_{min}) / (I_{max} + I_{min})$. The degree of polarization for emission and excitation is measured to be 93.2% and 55.4%. It indicates that the emission can be treated as the single dipole.

Figure 3-3. Emission enhancement of SPSs in GaN with a PSS structure. (A) Cross section SEM of the PSS structured gallium nitride. Cone shapes are the patterned sapphire structure. (B) The numerical simulation result of the far-field radiation pattern. The circles mark different collection half-angles which are from 100 to 900. (C) The numerical simulation result of the radiation pattern of the dipole in the gallium nitride with PSS structure. With the integration of all emission angle, we get that the intensity of the emission in C is two times higher than B. (D) Confocal scan map of the gallium nitride with PSS structure. The blue parts are the sapphire cones. (E) The brightness comparison between an SPS in a pristine gallium nitride (black squares) and an SPS embedded in gallium nitride with PSS structure. The saturated count for SPS with PSS reaches 2.33 Mcps (Mcounts per second) and on the contrary, the saturation curve in the pristine GaN is only 1.13 Mcps. The red solid curve is the fitting. (F) Comparison of count rate between pristine GaN and GaN with PSS structure with a fixed excitation power 10 mW. The shaded rectangles are guided to the eye.

3.1.3 Emission enhancement of SPSs in GaN with PSS structures

To further increase the brightness of the SPS, the mature fabrication techniques of GaN will favor the future applications. Another GaN sample is with a substrate which is called patterned sapphire substrate (PSS). First, we take a scanning electron microscope image of the structure as displayed in Figure 3-3A. The PSS structure is most widely used in the LED industry to enhance the extraction efficiency of the LED emission.

To demonstrate the enhancement effect of the PSS, we did a numerical simulation with 3D finite difference time domain (FDTD) with Lumerical. The simulation is carried out for both the pristine GaN and GaN on PSS structure [68]. For the pristine GaN substrate, the structure consists of a GaN layer whose refractive index is around 2.33 on top of sapphire layer with the refraction index of 1.75. For the GaN with PSS structure, the structure is modified to include the PSS structure. The numerical simulation result of the radiation pattern of an dipole sits in the pristine gallium nitride wafer and GaN with PSS structure is as shown in Figure 3-3B and Figure 3-3C, respectively. The circles mark different collection half-angles which are from 100 to

900. By comparing the total collected emission from the integration of the far-field pattern. We found that the PSS structure will induce an enhancement up to ~ 2 times.

The confocal scan map is as shown in Figure 3-3D. The red dot is the emission from SPE in GaN with PSS structure. We also measure the saturation curve for the SPS on a GaN sample without PSS and compared with the GaN with PSS. There is obvious emission enhancement and the SPSs located in the center of the GaN with PSS structure has saturation counts around 2.33×10^6 counts per second, compared to those in pristine GaN with 1.13×10^6 counts per second. We found that there is almost two times of enhancement for the SPSs with PSS structure compared to those without it. This also agrees well with the Lumerical simulation. Figure 3-3F shows a summary of the photo counts at a fixed power 10 mW in both the pristine GaN and GaN with PSS structure.

3.1.4 Origin of the SPSs in Gallium Nitride

Figure 3-4. Model to explain the wavelength distribution of the SPSs. (A, B)

Theoretical simulation of the distribution of the PL because of the different relative position between defect positions and cubic inclusions. The blue part shows different positions of the ZPLs and the arrows connect the different defect position with different PL energies. (C) If the point defects are exactly at the interface, it will result in two

different wavelengths 1100 and 1350nm ZPLs. In the real experiment, two PLs are also observed and it confirms the validity of our model. CBM, VBM is the conduction band and valence band respectively.

Finally, I will discuss the possible origins of the SPSs we found. Firstly, we rule out the possibility that the strain can generate such a big distribution of wavelength. We tentatively use a model of a point defect which is near cubic inclusions. We use a simple one-dimensional model to calculate the relative PL energies. Electron and hole recombination results in the emission. The hole is the defect which is very localized, but the electron is very sensitive to the potential. The relative position between the defect and the cubic inclusion will decide the exact wavelength. As the Figure 3-4A and B show the calculated distribution of the ZPLs from the cubic inclusions model. The model explains our experimental observations very well. Particularly, if the defect is sitting exactly at the interface between the cubic and hexagonal, it will result in two PL at around 1100 nm and 1350nm. Both are observed in the experiment [69-70].

Although the above model can help explain the distribution of wavelength found in our experiment, the exact origin of the SPSs are still unknown. There is no report so far to find any narrowband room-temperature GaN emission. If the GaN was implanted with Er ions, it has emission near 1.5 μm [71]. More investigations are needed to uncover the exact origin of the SPSs we found.

3.1.4 Complementary experiment

Figure 3-5. (A) Summary of PL of 10 SPSs in GaN in the telecom range. (B) Summary of ZPLs and FWHM of 18 SPSs in GaN.

Figure 3-6. Second order correlation measurements SPS 1. (A) CW $g^{(2)}(0)$ measurement at 4 different excitation powers, The black dots are the raw data and the red curve is the fitting. Inset figure is the zoom-in of the time between -20 ns to 20 ns. (B) Three energy level diagrams of the SPS1. To explain the bunching effect, a metastable state $|3\rangle$ must be included. k_{xy} represents decay rate from state $|x\rangle$ to state $|y\rangle$. (C-F) Three fitting parameters τ_1 , τ_2 , β , and k_{31} as a function of the excitation power. The black dots are the raw data and the red curve is the fitting with

the function $\frac{a}{\text{Exp}(bx)} + \frac{dx}{(x+c)}$.

To know more about the energy level of the SPS1, we performed a series of g^2 measurements at 11 different powers. I pick up four respective curves in Figure 3-5A and we can see that the bunching effect is obvious. To explain this bunching effect, a shelving state should be included in a three-energy level system. k_{12} is the excitation rate for the state $|1\rangle$ to state $|2\rangle$, this rate is proportional to the excitation power, ($k_{12} = \eta P$). Other are the rates from $|x\rangle$ to $|y\rangle$. The second order correlation can be written as:

$$g^2(\tau) = 1 - (1 + \beta) * \exp(-|\tau|/\tau_1) + \beta * \exp(-|\tau|/\tau_2)$$

Here β , τ_1 and τ_2 are

$$\beta = \frac{1 - \tau_2 k_{31}}{k_{31}(\tau_2 - \tau_1)}$$

$$\tau_{1,2} = \frac{2}{A \pm \sqrt{A^2 - 4B}}$$

where $A = k_{12} + k_{21} + k_{23} + k_{31}$, $B = k_{12}(k_{23} + k_{31}) + k_{31}(k_{21} + k_{23})$.

Experimentally, to get k_{xy} , we first obtain the value of β , τ_1 and τ_2 by fitting the data in Figure 3-6A with equation $g^2(\tau) = 1 - (1 + \beta) * \exp(-|\tau|/\tau_1) + \beta * \exp(-|\tau|/\tau_2)$. The values are shown in Figure 3-6C, 3-6D, 3-6E. The fitting agrees with the experimental data very well. Based on the fitting, the lifetime $(k_{21} + k_{23})^{-1}$ is around 776 ± 39 ps which is comparable the measured lifetime value 736 ± 4 ps measured by picosecond laser.

Figure 3-7. Model of SPS in gallium nitride with PSS structure. (A) Emission polarization pattern of SPS1, confirm that the SPS can be treated as a single dipole in the model. The blue dots are the raw data and the red curve is the fitting with $y = y_0 + A \cos^2(ax + \varphi)$. (B) Top view of the model, a dipole is locating at the center of three PSS cones. (C) Side view of the model, PSS structure are on the sapphire.

The model we built that to explain the emission enhancement of the PSS structure is as shown in Figure 3-7A. First, we showed the emission polarization of SPS 1. It is measured by varying the half-waveplate angle before the polarizer in the collection path, while the polarizer remains fixed. The figure indicates that the emission can be treated as a single dipole that sits in the GaN. Figure 3-7B and Figure 3-7C are the top and side view of our model in the Lumerical simulations respectively.

Figure 3-8. (A) Summary of PL measurements of 5 emitters in pristine GaN. (B)

Saturation measurement for these SPSs. For different emitter, the saturation raw data

is fitted well with the saturation equation as we mentioned above.

Figure 3-9. The photon correlation function of 5 SPEs in pristine GaN. The blue dots are the raw data and the red curve is the fitting. $g^2(0)$ is given in each graph.

Figure 3-10. (A) Summary of PL measurements of 5 emitters in pristine GaN with PSS structure. (B) Saturation measurement for these SPEs in GaN with PSS structure. For different emitter, the saturation raw data is fitted well with the equation we mentioned above.

Figure 3-11. The photon correlation function of 5 SPEs in GaN with PSS structure. The blue dots are the raw data and the red curve is the fitting. $g^2(0)$ is given in each graph.

To have a more detailed enhancement effect of the PSS structure. We found 5 SPEs each in pristine GaN and GaN with PSS structure. The PL spectrum and saturation counts are measured in each sample as Figure 3-8 and Figure 3-10 respectively. All emitters are confirmed to be single photon emission by g2 measurement as Figure 3-9 and Figure 3-11 displays. The saturation counts for the SPEs in pristine GaN are shown to be 0.5Mcps and in the GaN with PSS structure, it reaches up to 1.5Mcps which shows a distinct emission enhancement. All the SPE's lifetimes are summarized in Table 3-1. We found that the difference of the lifetime is not big between SPEs in GaN with and without PSS structure. So we rule out the possibility that the saturation counts difference is because of the modulation of the lifetime.

SPEs in pristine GaN	Lifetime (ps)	I_{∞} from saturation curve fitting	SPEs in GaN grown on PSS	Lifetime (ps)	I_{∞} from saturation curve fitting
A	484	0.89 M	a	482	1.4 M
B	448	1.13 M	b	442	1.17 M
C	348	1.67 M	c	405	2.13 M
D	456	0.89 M	d	348	2.33 M
E	265	1.19 M	e	396	2.45 M

Table 3-1. Summary of lifetimes and the saturation counts of SPE A-E in GaN without PSS and SPE a-e in GaN with PSS. The saturation counts are got from the fitting in Figure 3-8 and Figure 3-10. The table shows that the lifetime is similar between SPEs in GaN without PSS and GaN with PSS structure.

We calculated the ZPL position by using a simple quasi one-dimension model, in which a point defect locates close to cubic inclusion of three bilayers in wurzite GaN. PL emission comes from exciton recombination, in this process, the defect is acting as holes which is very localized. While the electron is loosely and will be very sensitive to the potential field which is generated by the cubic and hexagonal GaN bilayers. As a result, the energy will be related to the relative position between the point defects and the cubic inclusions.

3.1.5 Conclusion and outlook

To conclude, we present a high-brightness and stable single photon emission in the infrared range. The SPEs can exist at even room temperature and the FWHM is as small as 3 nm. Furthermore, we show that the emission can be enhanced to 2 or 3 times with the help of PSS structure which has been extensively used in the LED industry to greatly enhance the extraction efficiency. For the outlook, first, a more detailed model is needed to explain the origin of the SPEs and if some technique method can be used to generate the emitters like electron irradiation or ion implantation. Moreover, like those in the

NV center, if the electric excitation of the SPEs is feasible, and the p-i-n layer can be fabricated [72]. It is relatively easy to fabricate cavities or photonic waveguides in the GaN [73-74] and it is essential to get into the on-chip level [75-78]. Finally, to know if the emitter has spin properties is another important direction.

3.2 Bright room temperature single photon source at telecom range in cubic silicon carbide.

Single-photon emitters (SPEs) play a critical role in many quantum technology applications [58, 79-80] such as QKDs [64]. In such application, photons in the telecom range are preferred because of the minimum loss in the optical fibers. In this chapter, I will present a discovery of single-photon emission in the telecom range in 3C-SiC. The emission is stable and bright with MHz count rate at room temperature. Considering that silicon carbide's production and fabrication process are mature and advanced. Our work may pave the way to on-chip quantum technology applications.

3.2.1 Motivation of the investigation of SPSs in telecom range in 3C-SiC

Single-photon emitters (SPE) can be used as “flying qubits” to carry information and they are essential in many quantum technology protocols such as quantum technologies applications. For example, in QKD the key can be set out by encoding into the photons which is secure and protected by quantum mechanics. However, in most situations, the weak attenuated lasers have been used to demonstrate QKD experiments because of a true, bright and ideal for QKD is still missing. Photons from the single nitrogen-

vacancy center have been used to demonstrate quantum cryptography [81]. The most challenging thing is to find a room temperature SPE that is ideal for QKD which operates preferably at telecom wavelength. Photons lies in the telecom band will ensure minimum transmission loss in the optical fibers.

However, much efforts have been put to find SPE in telecom wavelength. One of the possible candidates is quantum dots. 120km secure QKD has been demonstrated already with QDs [34]. However, the most demanding thing is the cryotemperature which is not ideal for large-scale applications. Most recently, carbon nanotubes have been found to be a room temperature telecom wavelength SPEs hosts [30, 82].

SiC is a well-known semiconductor which is widely used in many areas, like LEDs, advanced high-temperature and high-power electronics. Recently, like nitrogen-vacancy center in diamond, different types of defects in SiC have been found to be able to optically addressed and coherently controlled [83]. For example, silicon vacancies[46, 84-87], carbon antisite–vacancy pair [12] [88]and divacancies [89-91]. All these emitters emit at visible spectrum range [92] or very weak in telecom range [90] which is not suitable for QKD applications.

In this chapter, I will present a new type of bright SPE with MHz repetition rate at room temperature in 3C-SiC. More importantly, the emitted photons lie in the telecom wavelength which ensures minimum transmission loss. We use the sample with a thin 3C-SiC epitaxy layer film which grows on its substrate. We found telecom wavelength

SPEs in 3C-SiC epitaxy layer and our results show that they are stable and bright, with Mhz repetition rate at room temperature. We investigate in detail the energy level with a set of second-order correlation measurements at different powers. Moreover, we measured the polarization pattern of those SPEs and find that they can be treated as a single dipole with nearly 97% degree of polarization which is ideal for polarization coding based QKD.

3.2.2 optical properties of SPSs in telecom range in 3C-SiC

The epitaxial layer on the Si substrate will introduce less unwanted background emission. What's more, getting thin membranes for photonics and micromechanics related applications is a more feasible way than bulk material. We measure the SPEs in 3C-SiC with our home built confocal microscopy system. The SPE was excited by a 950 nm diode laser through an oil objective with N.A. equals 1.35. The laser was purified with 1000nm short pass and in the collection path, the fluorescence from the emitters is filtered by 1000 long pass filter and then guided to single mode fiber which connects to the two-channel superconducting single-photon detectors (SSPD, Scontel) for a Hanbury-Brown and Twiss (HBT) setup for second order correlation measurement.

Figure 3-12a displays a confocal scan map of an area of $25 \times 25 \mu\text{m}^2$ on the epitaxy layer. All confirmed SPEs are marked with white circles. The nature of the single emission was confirmed with second-order correlation measurement. Next, we measured the PL spectrum of the SPEs. As shown in Figure3-12b, three SPEs's center wavelengths are 1085nm, 1188nm, and 1250nm respectively. It is clear that those SPEs have a PL distribution, thus we measure many SPEs's PL and summarize the center

wavelength of different emitters ranges from 1080nm to 1265 nm and the linewidth also varies around 150nm. Figure 3-12c shows the PL measurement of two SPEs at 4K which indicates that there is no obvious decrease of the linewidth. It implies that the linewidth broadening of the SPEs is not related to phonons. Please note that several sharp lines appear in low temperature might come from phonon coupling to the emission.

Figure 3-12. 3C-SiC single photon emitters. (a) The confocal scan of a $25 \times 25 \mu\text{m}^2$ area on the 3C SiC epitaxy layer with 2 mW 950nm laser excitation. All the confirmed SPEs are marked with white circles. (b) PL measurements of three representative emitters. (c) PL measurement of two representative emitters at cryogenic temperature (5 K). (d) Second-order autocorrelation measurement of SPE 4 with $g^2(0) = 0.05 \pm 0.03$. The excitation power is 0.2 mW. Black lines are the raw data and the red curve is the fitting. (e) Second-order autocorrelation measurement of SPE 4 with $g^2(0) = 0.13 \pm 0.02$ with 50 μW pulsed laser excitation.

Next, we perform the CW and pulsed second-order correlation measurement on one

of the bright emitters. Figure 3-12d displays the CW second-order autocorrelation measurement of SPE with 0.2mW 950nm excitation. The black is the fitting and the red curve is the fitting result. To minimize the influence of the unwanted background counts, the raw data $g^2_{\text{raw}}(\tau)$ was corrected by where $\rho = s/(s+b)$ [90]. Here, s and b are the signal and background counts, respectively. After the correction, it is fitted with

$$g^2(\tau) = 1 - (1+a)e^{-|\tau|/\tau_1} + ae^{-|\tau|/\tau_2}$$

where a , τ_1 , τ_2 are power dependent fitting parameters. We obtain $g^2(0) = 0.05 \pm 0.03$ From the fitting which proves its single photon nature. Figure 3-12e shows pulsed second-order autocorrelation measurement of SPE. The measurement was performed with a 950 nm picosecond laser. The repetition rate is 80Mhz. We obtained $g^2(0) = 0.13 \pm 0.02$ with the integration of all the area under each peak. Which also proves the emitter is indeed SPE.

To know more the optical properties of the SPEs we found. We measured the lifetime of SPE 4 using a picosecond laser with 80 MHz repetition rate. Figure 3-13a, 3-13b shows the lifetime and PL measurement of the SPE 4. A single-exponential equation $I(t)\exp(-t/\tau)$ where $I(t)$ is the fluorescence intensity, and τ is the fluorescence lifetime is used to fit the decay which displays in Figure 3-13a. From the fitting, lifetime of 0.81 ± 0.01 ns is obtained. Following the lifetime measurement is the saturation power monument. We vary the excitation power and record the emission counts at different excitation power which has been shown in Figure 3-12c. The black part is the raw data and the red curve is the fitting with $I(P) = I_s / (1 + P_0 / P)$, in which I_s , P_0 is the saturation counts and saturation power, respectively. From the fitting, we obtained that I_s is 1.36 Mcps. P_0 was about 3.0 mW. Finally, we care about the fluorescence

stability of the emitter, we measured the fluorescence live count trace of the SPE4 with 1.5 mW and 8.2 mW excitation respectively. The bin size is 100 ms. The duration is 400s. AS is displayed in Figure 3-13d. No obvious blinking or bleaching has been observed even at very high power which verified that the SPE is a room temperature photostable single photon emitter.

Figure 3-13. Single photon emitter characterization. (a) The lifetime of the SPE. The black curve is the raw data and the red solid curve is the single exponential decay fitting $I(t)\exp(-t/\tau)$. From the fitting, we got the lifetime of the SPE is $\tau = 0.81 \pm 0.01 \text{ ns}$ (b) Room temperature PL measurement of the SPE 4. (c) Count vs power measurement to know the saturation power. The black dots are the raw data and the red solid line is the fitting with $I(P) = I_s / (1 + P_0 / P)$. (d) Fluorescence stability measurement at different laser powers 1.5 mW (black) and 8.2 mW (blue) respectively, we haven't observed any blinking behavior.

Three-level model

To understand more photophysics of the emitter, we measure second-order correlation under a set of excitation powers, and we include a three-energy level model[12, 84-85, 88] to explain what we have observed. We pick up three $g^2(t)$ measurement are shown in figure 3-14b. The red line is the fittings of the data. From the figure, an obvious bunching effect has been observed which could be explained with the help a three every level model involving a metastable state. Figure 3-14a shows the model with a ground state $|1\rangle$, and excited state $|2\rangle$ and metastable state $|3\rangle$. Also, different excitation and emission rate have been marked in the Figure. Figure 3-14 c-e displays the relationship between fitting parameters a , τ_1 and τ_2 and the power. a is most related to bunching effect, and τ_1 can tell the transitions rate between the ground and excited state, τ_2 is more related to the metastable state.

Figure 3-14. Photophysics analysis for SPEs. (a) The three-level model is used explain the observed bunching effect. (b) Three continuous wave second-order correlation measurements with a set of laser excitation powers. The red curves are the fitting (c), (d), (e), The fitting parameters a , τ_1 and τ_2 with respect to the power.

We fit the parameters based on the following equations[16]:

$$a = \frac{1 - \tau_2 k_{31}}{k_{31}(\tau_2 - \tau_1)},$$

$$\tau_{1,2} = \frac{2}{A \pm \sqrt{A^2 - 4B}},$$

In which the $A = k_{12} + k_{21} + k_{23} + k_{31}$, $B = k_{12}(k_{23} + k_{31}) + k_{31}(k_{21} + k_{23})$. Which agrees with the experimental data very well. Moreover, the calculated lifetime, $(k_{21} + k_{23})^{-1}$, 0.7 ± 0.05 ns is comparable to the measured lifetime value of 0.81 ± 0.02 ns.

3.2.3 Polarization pattern of SPSs in 3C-SiC

Figure 3-15. The polarization pattern of the SPEs. (a), (b), polarization pattern measurements of one emitter for the excitation and emission respectively. The dots are the raw data and the solid curves are the fitting with $I = a + b\sin^2(\theta + \phi)$. (c) Emission polarization of 12 emitters, it indicates that there is none preferable emission axis.

To figure out if the emitter has a single dipole or multi dipoles, we investigate the excitation and emission polarization pattern by exciting the emitters with linear polarization excitation laser beam. The polarization of the excitation beam is realized by rotating a half-wave plate after the polarizer. Figure 3-15a and 3-15b are the excitation (blue) and emission (red) polarization pattern measurements of one emitter. The dots are the raw data and the solid curve are the fitting with $I = a + b\sin^2(\theta + \phi)$, in which a, b, θ and ϕ are the free parameters of the fitting. The degree of the polarization which is defined by $\eta = (I_{\max} - I_{\min}) / (I_{\max} + I_{\min})$, the excitation polarization degree is 97% and emission polarization is 98%. We found that all emitters we found have the degree of polarization for both the excitation and emission that are more than

90 %. It confirms that the emitter has a single dipole transition. Figure 3-15c shows the summary of the polarization axis of 12 emitters we found. No obvious preference of the emission polarization has been observed. The high polarization degree higher than 95% emission indicates that they are ideal SPEs for QKD scheme with polarization coding, for example, the Bennett-Brassard protocol.

3.2.3 Discussion of the origin of the SPEs in 3C-SiC

In this section, I will discuss the origin of the SPE we found. First, we conclude that those emitters we found with similar properties, but different center wavelength are same origin for the following indicators: First, the SPEs' emission spectrum are the similar at both room and 5K. Moreover, they have similar linear polarization as single dipoles and the observed similar bunching effect. To confirm that the SPEs are indeed in the 3C-SiC epitaxy layer not in the Si substrate, we use reactive ion etching to etch the samples with 1 μ m epitaxy layer. The three samples are etched 200 nm, 300 nm, and 1 μ m depth respectively. In the sample which has been etched for 200nm and 300nm, we found the SPEs successfully, however, no SPEs have been found in the sample etched with 1 μ m which confirms that the SPEs are in the epitaxy layer, not related to the Si substrate. Moreover, we also perform electron irradiation of the samples with 1Mev electrons at a range of fluences from $1\text{E}15\text{cm}^{-2}$ to $5\text{E}16\text{ cm}^{-2}$. After the irradiation, the samples were then annealed in 800 degrees for 30min with high vacuum. Next the sample was annealed with 800 degree in high vacuum atmosphere in high vacuum for 30 min. We found that neither the electron irradiation increases the density of the emitter nor new emitters have been created as Figure 3-16.

Figure 3-16: Electron irradiation of 3C-SiC: Confocal scan map of the electron irradiated 3C-SiC sample. No obvious effect like emission enhancement or defect density increment has been observed.

As shown in Table 3-2, besides the sample with 1 μm epitaxy layer, we have also tested another wafer with 2.7 μm epitaxy layer, which is also on Si substrate and purchased from Air Wafer Inc. We found the Similar SPEs which shows that the SPEs are common and reproducible as Figure 3-17 shows.

	Sample A	Sample B
Epitaxy layer?	yes	yes
Epi side surface orientation	(111)	(110)
Epi thickness	1 μm	2.7 μm
Doping	No intended doping	No intended doping
N type?	yes	yes
Substrate (thickness)	Si (1000 μm)	Si (725 μm)

Table 3-2: Two different samples have been tested.

Figure 3-17: SPEs found in sample B a. PL measurement of 2 repetitive SPEs in sample B. b. Second-order measurement to confirm they are single photon emission.

From the comparison with the emitters in the visible range which have been reported already. We found that they are different. In the infrared range, one of the SPEs in 3C-SiC which has been reported before is the divacancy defect which is called Ky5/L3 defect. However, it has a fixed and narrow zero phonon line at low temperature. Moreover, another major difference is the brightness. The photon counts reported in several pieces of literatures are tens of kilo counts which are far less our emitters. The emitters found have a variable center wavelength distribution which is more than 100nm difference. Strain unlikely can introduce such big difference and like SPEs in gallium nitride which has been reported recently, we infer that our emitters PL variance

could be also induced by stacking faults in the 3C-SiC different layers. The photoluminescence is the result of electron-hole recombination. The defect is very localized, and the electron is loosely localized which is very sensitive to the potential field. Thus, the zero-phonon line position is determined by the relative position between the defect and the inclusion. We have also done the PLE measurement by varying the wavelength of the excitation laser beam. As shown in Figure 3-18, for the emitter whose ZPL is around 1275nm (0.97 eV), the most efficient wavelength is around 975 nm (1.27 eV).

Figure 3-18: PLE measurement of the SPE: The excitation wavelength of the SPE varies while the power remains constant at 0.5 mW. The black curve is the raw data off-resonant PL spectrum of the SPE which is fitted with Lorentz fitting. From the fitting, linewidth can be extracted which is around 181 ± 3 nm. The energy level we introduce to explain it is inserted to the middle.

Refer to the Franck-Condon principle, the peak of the PLE spectra will be blue-shifted with respect to its ZPL position, and because of the complex form of the vibronic bands, it is usually asymmetric. The energy level diagram is as shown in the insert figure, it

involves one ground state E_g and two excited level E_1, E_2 . First off-resonant laser pumps the emitter from the ground state to E_2 . Then the emitter will to E_1 . Finally, it radiatively returns to the ground state and emits photons.

To conclude, I present the observation of single photon emission found in 3C-SiC epitaxial films which emit at telecom wavelength. These SPEs are photostable and bright, in the order of MHz repetition rate. Several immediate directions are as follows: The emission could be possibly enhanced further with photonics structures like solid immersion lenses[84], pillar structures and photonic crystals[93-94]. Moreover, it is interesting to see if the SPEs can be electrically excited [72] and it will benefit the large-scale integration and on-chip quantum technologies.

3.3 Direct writing of single germanium vacancy center arrays in diamond

Solids state quantum emitters in diamond are promising to be qubits (building block of quantum computation) for many quantum technology applications[4, 52, 95]. SPEs with inversion symmetry is preferable because of their superior optical properties. In this section, I will present a method to generate SPE array in diamond with the technique of maskless implantation of germanium vacancy (GeV) centers. With direct and tens of nanometer precision implantation, single GeV arrays are engineered. More importantly, the single photon emitter generation ratio reaches up to 53% in 200 Ge⁺ dose area. This method provides a platform for the investigation of spin-spin

interactions with GeVs and also will benefit future Nanofabrications.

3.3.1 Germanium vacancy in diamond

SPEs in solid-state are promising building blocks of many quantum technologies applications. Especially those with superior optical properties are preferable. One of the ideal optical properties is narrow bandwidth emission like high Debye-Waller factor. Much efforts have been put into searching a good candidate. Among them, the most well-known and studied one is nitrogen-vacancy center[15, 96]. Very recently, silicon vacancy (SiV) in diamond have attracted increasing attention due to the very high Debye-Waller factor, nearly 80% of its emission lies at its own zero phonon line[16, 97-99]. However, one disadvantage is its short coherence time. SiV has around 40 GHz ground state, the single-phonon absorption from the lower to the upper branch will more likely to happen[23]. Thus, a larger ground state splitting which will suppress the process is preferable. Germanium vacancy (GeV) is a promising one. Because of the similar atomic structure, GeV also has similar Debye-Waller factor with SiV[17, 100]. Moreover, it has a larger ground state splitting ~ 120 GHz, almost three times as SiV.

3.3.2 Fabrication of single GeV array with ion beam (FIB) implantation technique

Here, I will present a direct scalable fabrication of single GeV technique. The precision of this method can reach up to tens of nanometers and the single photon emitter generation can reach up to 53%.

Figure 3-19. GeV ensembles generated by FIB ion implantation. (a) Atomic structure of the GeV in diamond. (b) Depth simulation of 35 keV Ge ions implanted into the diamond by using SRIM. The inserted figure is 500 ion's trajectories from the simulation. (c) Implantation pattern from initial design, the two lines are the ensembles areas whose dose is high. The four different colors represent different doses of Ge ions into the spot. The right figure is the confocal scan map of 4 different doses. (d) PL measurement of the two lines where GeV ensembles are created at room temperature, its ZPL is around 602 nm. (e) PL measurement of the GeV ensembles at 4 K, the signature transitions A B C D is labeled, and ground state splitting is around 184 GHz and excited state is around 1139 GHz.

Figure 3-19a displays the atomic structure of the GeV defect. Germanium atom replacing two adjacent vacancies in the diamond lattice forms a GeV. The structure of GeV is similar to the SiV, thus GeV also has a D_{3d} symmetry which is not sensitive to the surrounding fields. A 35 keV nanoFIB system (ionLINE, RAITH Nanofabrication) is used to generate the GeV. An extremely focused Ge ion beams within a few nanometers spot size on the sample. This ensures the tens of nanometer precision. From the monte carlo simulation, the 35 keV Ge ions will penetrate the diamond for around 25 nm (Figure 3-19b). The longitudinal and lateral position uncertainty is around 5 and 4 nm. Also the ion beam size is roughly 40 nm, the spatial resolution we estimate

can reach is around 60nm in three axes[54, 101-102].

The bulk sample we used to implant is a commercially available CVD growth diamond purchased from Element Six $[N] < 1$ ppb. We implanted four different doses from 100 to 700 Ge ions by adjusting the beam current and the dwell time of the FIB system. The designed figure is as shown in Figure 3-19c, two perpendicular lines are the ensemble areas with 700 Ge⁺ ions implantation. Four different colors represent different implanted doses. Spots are separated by 2 μ m for two directions. After the FIB ion implantation, the sample was then annealed at 1000 °C for half an hour.

The FIB ion implantation result is characterized by a home built confocal microscopy system. The emitter is excited by a 532 nm diode laser. The emission is guided to a spectrometer for PL measurement or APD for counting the photons. Figure 3-19c displays a scanned map from our home built confocal microscopy system of the 4 areas with different implantation doses. To confirm what we have created is indeed GeVs, we have performed PL measurement both at room temperature and at 10K as shown in Figure 3-19 d and Figure 3-19 e, respectively. At 10 K, the PL is the characteristic ABCD transitions of GeV center. The origin of the ABCD can be explained by the inset of Figure 3-19e. Due to the spin-orbit coupling, the ground state and excited state will split. From Lorentz fitting, the ground splitting are 184GHz and the excited state splitting is $\Delta e = 1139$ GHz, respectively which is comparable to previous literature.

Figure 3-20. Single germanium vacancy center in diamond. (a) The scan of the 200 Ge⁺ area.

single GeV is marked with a white circle. (b), (c) g² measurement for both a single GeV center and two GeV centers, the $g^2(0)$ is fitted to be 0.15 and 0.57 respectively. (d) Saturation behavior measurement of the SPE A, the black was the raw data and the red solid curve is the fitting with $I(P) = I_{\infty}P / (P + P_{sat})$. (e) The lifetime of the SPE with 532 nm pulsed excitation, the raw data is fitted with simple single exponential decay. We obtained the lifetime $\tau = 4.1 \pm 0.2 \text{ ns}$. (f) The photostability measurement with 0.1s time bin and 2 min duration at the excitation power 2.8 mW and 6 mW respectively.

3.3.3 Conversion yield and single photon generation ratio analysis

Next, in the 200 Ge⁺ implanted area, we found single germanium vacancy center. Figure 3-20a, it is marked with white circle. To confirm it is indeed single photon emitter, a second-order correlation function, $g^2(\tau)$, the backgrounds was filtered out by a 600 nm band pass. The raw data $g^2_{\text{raw}}(\tau)$ was corrected by where $\rho = s/(s+b)$. s and b are the counts for signal and background respectively[103]. The background was got by moving the excitation 2 micrometers away from the SPE. The black are the

backgrounds corrected fitted with $g^2(\tau) = 1 - \alpha * \exp(-|\tau|/\tau_1) + \beta * \exp(-|\tau|/\tau_2)$. We obtained $g^{(2)}(0) = 0.15 \pm 0.05$ which is far below 0.5 which confirms it is single photon emitter. Figure 3-20c shows an example of second order autocorrelation of two GeV emitters, $g^2(0) \sim 0.57$ was obtained from the fitting.

To uncover more for the photophysical properties of the implanted GeV, we measured saturation behavior of the SPE and lifetime of the excited state. As displayed in Figure 3-20d, the black dots are the raw data and the red solid curve was the fitting with $I(P) = I_{\infty} P / (P + P_{sat})$. The lifetime of the excited state is measured to be $\tau = 4.1 \pm 0.2 \text{ ns}$ from a single exponential decay. The photo stability was measured at different excitation power 2.8mW and 6mW respectively with 0.1 s time bin and 2 min duration.

Figure 3-21. (a) Analysis of the number of GeV centers per spot, total number is 90. (b) Conversion yield of the 4 different implanted doses.

Next we analyze the Ge ions to GeV center in diamond, first, we fix the photon counts for a single and two GeVs and then normalize all the spots based on it. Then calculate and estimate how many emitters we have in each spot. For each dose, we have scanned 90 spots. The confocal maps are displayed in Figure 3-19c (right). Figure 3-21a shows the summary of the number of emitters for every spot of 4 different doses. In 100 GeV

area, 10 of the 36 spots are dim without any emission detected, no emitters generated. When the dose of the implantation increases from 100 Ge⁺ to 200 Ge⁺ each spot, single GeV centers are dominated. The single photon emission ratio is estimated to be 53%. If the dose still increases from 200 Ge⁺ to 400 Ge⁺ or even 700 Ge⁺ each spot, most likely the spots turn to two emitters and even three emitters. Finally, we calculate the conversion yield by the average number of emitters each spot for each dose over the total ion number as shown in Figure 3-21b. We found that the conversion yield remains around 0.6% with the dose from 100 Ge⁺ to 700 Ge⁺. We found that this conversion yield is very similar to silicon vacancy in silicon carbide [103] and in diamond [104], and comparable with nitrogen implantation [105-106]. We have summarized the previous single photon generation ratio and conversion yield of SiV in diamond, SiV in silicon carbide and our result as displayed in the Table 3-3. We found that the highest conversion yield is about 15% for SiV in diamond. The conversion yield for GeV in diamond is low compared to others 0.4-0.7% but the single photon generation ratio can reach up to 53%. This is extremely helpful in the deterministic fabrication of the GeV center with high spatial resolution. Please note that the conversion yield is possible to be further enhanced by improving the annealing temperature [101].

	SiV in Silicon Carbide [20]	SiV in Diamond [21]	GeV in diamond (our work)
Emitter conversion yield (%)	1.4-4.2	15	0.4-0.7
Single photon generation ratio (%)	38	13.3	53.33

Table 3-3. Summary of three different SPEs' conversion yield and single photon

generation ratio.

3.3.4 Conclusion and outlook

To conclude, I present a deterministic fabrication of single GeV center arrays in diamond. This method can reach tens of nanometer spatial resolution and there is no need to include the mask. Although now the conversion yield is relatively low, it can be further increased the future, using the method like high-temperature annealing and electron irradiation. Our technique will benefit a lot of quantum technologies applications with GeV centers like coupling the defect to many photonic structures such cavities [107-109] and waveguides [110]. This method can also favor fabrication GeV centers, which may involve the coupled electron spins with entanglement between spins [111-112].

Chapter 4 Spin properties investigation: coherent control of silicon vacancy optical transition in diamond

In the realm of quantum information processing, whether a system can be prepared, read out and finally coherent control is of great importance. Scientists always prefer to develop an on-chip quantum platform which involves both quantum nodes and coherent photons. The optical transition of the quantum emitter will exhibit many theoretically predicted phenomenon such as dressed states and Autler-Townes splitting. In this chapter, I will introduce our work on the coherent control of spins in silicon vacancy in diamond and based on the optical transitions in the silicon vacancy center in diamond, we excited the transition with an intense laser field and study the light-matter interaction in the process. Especially, we successfully control the spin states of the transition coherently.

4.1 Silicon vacancy in diamond

Coherent control of spin states of the quantum emitter is of the central importance in the quantum information processing[112-116]. Especially, solid state spins and photons can interact with each other which provide an important platform for various of quantum technologies[113-114, 117]. Many types of solid-state defects have been extensively studied so far. Some of them show excellent both optical and spin properties. Among them, the most extensively studied one is the nitrogen-vacancy centers (NV) in diamond. The spin photon and spin-spin entanglement have been demonstrated successfully[113, 117]. Nevertheless, NV is not a good option for the following two

reasons: first, the zero-phonon line (ZPL) only consists 3-5% of the total fluorescence. Also, there is strong inhomogeneous broadening of the emission which limits the coherence of the photons. Very recently, another emerging interesting single photon emitter is the silicon vacancy center which has excellent optical properties owing to its unique inversion symmetry. With this symmetry, the optical transitions are more robust against the environmental fields which are better than NV centers. Moreover, around 80% of the photons lines in the zero-phonon line and in the low-strain bulk diamond, the nearly lifetime-limited linewidth transitions have been found[16, 21, 118-122]. At the temperature of 4K, the coherence time is around 40ns[123-124]. If the temperature is further decreased to millikelvin, the coherence will be significantly enhanced because now the coherence time is only limited by the spin relaxation time[125]. To conclude, silicon vacancy has become a very promising quantum qubit and a quantum node in the quantum information sciences.

Preparation and coherent control of the transition of the silicon vacancy centre are of great importance. If an ideal two-level system is excited with an intense laser field, the population of the state will vary between the ground and excited states, which is also called Rabi oscillations. If we transfer the optical Rabi oscillations from the time domain to the frequency domain when the excitation laser is the continuous wave (CW) laser beam. The whole system will show Mollow triplet spectrum[126], which indicates that coherence control has been achieved and it is also a single photon generation method with the frequency away from the resonance[127-135]. In the work, a transition is been strongly driven and can be coherently controlled with laser field. With both pulsed laser and CW laser field, the optical Rabi oscillation has been observed. Fourier

transform of the optical Rabi oscillations enables us to observe Mollow triplet spectrum. Moreover, the Autler splitting has been also observed based on the two-laser scan scheme. The coherence of the photons has been identified in both high and low power regime.

The silicon vacancy center is formed by a silicon atom taking the place of two adjacent carbon vacancies[136] as displayed in Figure 4-1a. SiV is negatively charged and the symmetry of it is D_{3d} which is robust the environment fields [137-139]. The energy levels and optical properties have been reported in [97, 138-139]. Because of spin photon coupling, the silicon vacancy centre exhibits ABCD four zero phono lines in the low temperature as shown in the Figure 4-1b. We resonantly excite one of the transitions as displayed in the Figure 4-1b, the transition C is excited. There is no fast relaxation from the upper branches, so the transition C will exhibit more count rate and longer coherence than other 3 transitions. As shown in the Figure 4-1c, we only collect phonon sideband photons more than 750 nm and a very narrow PLE spectrum is shown in Figure 4-1c. The laser was scanned, and the wavelength was recorded from wavelength meter (Highfinesse WSU-30) with maximum resolution of 5MHz. The linewidth is around 219 MHz which is very close to the lifetime limited linewidth 86MHz. The lifetime was measured as Figure 4-1d shows.

Figure. 4-1: Silicon vacancy center. **a.** Schematic structure of SiV in diamond. **b.** The energy level diagram of the typical SiV with excited state splitting $\Delta_e = 410\text{GHz}$ and ground state splitting $\Delta_g = 228\text{GHz}$, respectively, A, B, C, D transitions are marked. If transition C is excited with an intense laser field, and the dressed states are labeled as $|g,n\rangle$, $|e,n-1\rangle$, $|g,n+1\rangle$ and $|e,n\rangle$. **c.** The Resonant excitation spectrum of the transition C. **d.** Lifetime measured with ~ 100 ps laser pulse.

4.2 Optical Rabi oscillation in SiV

The optical Rabi oscillation is measured with a superconducting nanowire detector with quantum efficiency around 60%. The two-level transition is excited with an intense laser pulse, which yields the oscillation of the population of the excited states. The population can be expressed by $\sin^2(\Omega_g t / 2)$, in which $\Omega_g = \sqrt{\Delta^2 + \Omega^2}$ and Δ is the detuning of the laser, $\Omega = \mu E / \hbar$ is Rabi frequency, μ is the dipole moment of the optical transition. E is the amplitude of the driving field. While if we see it in the frequency domain, the dressed state can be included to explain it very well as shown in

figure 4-1b (red dashed lines). Both the ground and the excited states will split into two states which are separated by an energy $\Omega_g = \sqrt{\Delta^2 + \Omega^2}$.

The silicon vacancy transition is excited with a 5 ns laser pulse. The fluorescence was recorded by APD and photon correlation measurement system. As displayed in Figure 4-2a, the frequency of the optical Rabi oscillations is higher with more intense excitation laser field. The curves are fit with [129]

$$P = 1 - e^{-\eta|\tau|} \left(\cos(\mu|\tau|) + \frac{\eta}{\mu} \sin(\mu|\tau|) \right)$$

Here $\eta = 1/2T_1 + 1/2T_2$ and $\mu = \sqrt{\Omega_g^2 + (1/2T_1 - 1/2T_2)^2}$ with T_1 (lifetime) and T_2 (photon coherence time) respectively. T_2 and Ω_g are the free parameters in the fitting while the measured T_1 value is $1.85 \pm 0.02 \text{ ns}$. As displayed in Figure 4-2b, the extracted frequency of Rabi oscillation is linearly proportional to the square root of the laser power. The average T_2 is around 1.62 ns which is smaller than the ideal value $2T_1 = 3.7 \text{ ns}$. It may be due to the pure dephasing of the excited state. Moreover, as displayed in figure 2c, T_2 is more than 1.3 ns even with 12GHz Rabi frequency which is excited with 300 times saturation power. No obvious power induced dephasing is observed. This robustness could be related to the insensitivity to the electric fields [21].

Figure 4-2d shows the intensity profile with the respect to the laser detuning. If a detuned laser is added, the Rabi frequency will be generalized to $\sqrt{\Omega^2 + \Delta^2}$. The Fourier transform of the curve is shown in Figure 4-2e. The dashed lines in the Figure 4-2e agrees well with $\nu_0 + \Delta \pm \sqrt{\Omega^2 + \Delta^2}$ with Ω extracted in the 0-detuning case.

Figure. 4-2: Time-resolved optical Rabi oscillation. **a.** Three different optical Rabi oscillations of C transition. The period of the laser pulse period is 15 ns. The dots are the raw data and the solid curves are the fitting of the curves with equation (1). From the fitting, the Rabi frequencies are $\Omega/2\pi = 0.906\text{GHz}, 1.304\text{GHz}, 1.854\text{GHz}$. **b.** Fitted Rabi frequency is linear with the square root of the excitation power. **c.** The extracted T_2 with respective the different powers. The value of T_2 stays above 1.3 ns even with Rabi frequency of 12 GHz corresponding to 300 times of saturation power. **d.** Detuning of the excitation laser and the intensity profile. **e.** Fourier Transform analysis of the **d**. In the frequency domain Mollow triplet is shown clearly. Two side branches agree well with the theoretical values $\nu_0 + \Delta \pm \sqrt{\Omega^2 + \Delta^2}$.

4.3 Correlation measurement in SiV

To know more about the quantum behaviour of a single SiV defect, we measured a series of second-order correlation function at different excitation power with a Hanbury-Brown-Twiss (HBT) setup. As shown in Figure 4-3a, the resonantly excited SiV exhibits a $g^{(2)}(0) = 0.061 \pm 0.026$ with the saturation power. When the excitation

power is stronger, oscillations are clearly seen in the time range of $t \in [0, \pm 5] \text{ ns}$. When the oscillation is measured by the $g^{(2)}(\tau)$ measurement. The start signal is when the detector detects the first emitted photon. The following photon indicates the population of the excited states. The measured $g^{(2)}(\tau)$ is then fitted with

$$P = 1 - e^{-\eta|\tau|} \left(\cos(\mu|\tau|) + \frac{\eta}{\mu} \sin(\mu|\tau|) \right)$$

and the extracted Rabi frequency is linear with the square root of the excitation laser power.

Figure. 4-3: Photon correlation measurements. **a.** Second order correlation function $g^{(2)}(\tau)$ measurement in different powers, the black part is the raw data and the red solid curves are the fitting. **b.** The extracted Rabi frequency is linear with the square root of the excitation laser power.

4.4 Autler-Townes splitting

Apart from the Fourier transform of the real-time optical Rabi oscillations, in the frequency domain the Autler splitting can be observed. We excite the C transition with strong laser power and another laser is scanned across D transition. The scanning results are as shown in Figure 4-4a. The system can be regarded as a Λ system with one

transition C to be heavily pumped ($\Omega_C^2 \gg P_{sat}$) and transition D is weakly pumped. C and D's excited states are the same, but ground states are different. A laser is tuned to transition C and another laser is scanned over D.

Very similar to the well-known coherent population trapping (CPT), the photon emission will have a dip around $\Delta_D = 0$ due to the destructive interference between C and D. And if the transition C is strongly pumped and the splitting of the scan image can be regarded as Autler-Townes splitting.

At the same time, we change the intensity of the resonant laser which in the C transition and remain the D transition power. Figure 4-4b shows the normalized photon counts with respect to the laser detuning Δ_D . We can obtain the Ω_C from the fitting function with Lindblad master equation. We found that if the excitation power increased, the dip width also increased. From the different power fitting, Ω_C is linear with the square root of power (Figure 4-4c), which agrees with the previous results in our optical Rabi oscillation measurement. We performed further measurements in which both C and D transitions are both detuned, and the measured data is as displayed in Figure 4-4d and simulation with Figure 4-4e.

Figure. 4-4: Detection of Autler-Townes. **a.** The energy level of the system, a strong laser is applied in the transition C while another weak laser is scanned across D transition. **b.** The transition C is excited with three different powers and the photon emission from the transition D is recorded $[-0.7GHz, 0.7GHz]$ from resonance. **c.** Extracted Ω_C is linear with the square root of the excitation power. **d.** The transition C was detuned $[-1.5GHz, 1.5GHz]$ from resonance while the transition D is detuned from resonance $[-1.5GHz, 1.5GHz]$, the photon intensity profile is recorded. **e.** Simulation data in the **d**

Figure. 4-5: Fine structure of silicon vacancy in diamond.

In the nanodiamond. We found there are many optical transitions that belong to different SiVs. How to distinguish those transitions and filtered out four optical transitions that belong to the same single silicon vacancy center is a challenging thing. We resonantly excited on of the transition with little power and measure the PL spectrum to see if other transitions can be detected. As shown in Figure 4-5, when we pump A, the PL near A is saturated which is reasonable because we haven't added any long pass filters the weak laser was guided the spectrometer directly. However, more importantly, other transitions like C,D are clear. Based on the same method, we excite one of the transitions and detected other transitions, all four transitions that belongs to the same single SiV has been figured out.

4.5 Conclusions and outlooks

To conclude, we successfully achieved coherence control of one transition in the silicon vacancy in diamond. The silicon vacancy exists in the nanodiamond particles. It has several advantages over those in the large ultra-pure bulk crystal. For example, the diamond which hosts the silicon vacancy can be put into the photonic crystal cavity or waveguide. Moreover, the coherence time remains even at the very high excitation power. Which is a very good property for the quantum nodes connection with Hong-Ou-Mandel interference experiments [54]. Also, several interesting in the future is the photons from the Mollow triplet may be filter out with the help of Farby-Perot filters [140] and Michelson interferometers. The triplet emission can be studied further [140]. Moreover, the Mollow triplet sideband photons can be coupled to the optical cavity which can be greatly enhanced [141-142] and also the tunable dressed state laser is possible [142]. Finally, if there is a magnetic field, the spin will split in the Mollow triplet which is helpful to spin-photon interface or single-shot readout [135].

Chapter 5 Application of solid-state SPSs: Thermometry with infrared photons and defect spins in silicon carbide

High sensitivity and high spatial resolution are two important parameters. New emerging “quantum sensors” which are the defect spins in solids have drawn much attention for their superior properties. Moreover, if the spin sensor in the solids is robust against environment noise is also an important parameter. In this chapter, I will present a high spatial resolution and high-sensitivity thermometry with defect spins in silicon carbide (SiC). The thermometry was demonstrated with the Ramsey based thermometry. This work may provide a thermometry scheme that can reach high spatial resolution and high sensitivity. It will be helpful in some real thermometry in microelectronic or biological systems.

5.1 Thermometry with divacancy spins in 4H SiC

In modern society, to find a precise, high spatial resolution and noninvasive thermometry method has motivated researchers in many areas to achieve this goal [143]. Particularly, in the biological or physical research [143-144], a quantum sensor with sub degree and high biocompatibility is always preferable. To this goal, several systems have been found to be suitable for the thermometry, such as Raman spectroscopy [143, 145], quantum dots [143, 146], organic dyes [143, 147] and scanning thermal microscopes. However, there are many limitations in those methods such as blinking and bleaching of the fluorescence, low sensitivity, also many errors because of the fluorescence rate fluctuations [51, 143-144]. To overcome these drawbacks, the solid-state spins in the defects act as “quantum sensors” have been developed. Among them,

the most studied one is based on the nitrogen vacancy (NV⁻) center in diamond [148-150].

Usually 532 nm green laser is used to excite NV center and its emission photons is from 600 to 800 nm. However, there is a disadvantage of this as light at such wavelengths will induce more optical damage to living biological system [151-152]. Moreover, for usually there is a very noisy environment in the biological systems, a self-protected thermometry method which enables the system robust to the environment noise is preferable. So that's the motivation of us to develop a thermometry method which is robust to the environment, has a self-protected mechanism. More importantly, the photons from the neutral divacancy defects spin in 4H-SiC are in IR spectral region which will cause less damage to the biology systems [91, 153-157]. Another advantage of defects in 4H-SiC is that SiC is well known semiconductor whose production and engineering technique are mature in the realm of electronic and electromechanical devices [153]. Another advantage is that NV center has 4 orientations [15] and it will result in a decrease of the sensitivity. One type of the spins in the divacancy has the exact same orientations and in terms of the sensitivity, it will improve a lot. The ground state of the neutral divacancy has the spin-1 and 6 different forms of the divacancy both in 4H-SiC and 6H-SiC (named PL1-PL6 and QL1-QL6) [91, 153]. The spins in the SiC have several excellent optical properties, such as fabrication friendly, good biocompatibility and extremely long coherence time (~ms) [91, 153, 155-157]. And many quantum operations have been demonstrated such as quantum sensing and coherent control of the spins [91, 155].

In this chapter, I will talk about a self-protected thermometry method with the

divacancy spins in 4H-SiC. Our thermometry method is based on the Ramsey fringes of the electron spins in 4H-SiC. We found that the spins are very robust to the environment noise and we attribute it to a self-protected mechanism which protects the spin from the fluctuations around the spin. We also investigated the change of the optically detected magnetic resonance (ODMR) with respect to the temperature (20 K to 300 K). The zero-field splitting D is linear with the temperature. And this linear relationship is the base for our thermometry. Finally we successfully demonstrate the self-protected thermometry with infrared photons with the Ramsey fringes method. The sensitivity of our thermometry can reach up to $200 \text{ mK/Hz}^{1/2}$. Our method is extremely useful in the microelectronics and biological applications.

5.2 Theoretical models for the thermometry with spins

Firstly, we marked the spin-1 system of the PL5 [91, 153] as ($|\uparrow\rangle, |0\rangle, |\downarrow\rangle$). When the magnetic field is added to the system, the ground state will split, and it will result in two ODMR resonance at $D + E$ and $D - E$. The spin Hamiltonian can be expressed as

$$H = H_0 + H'$$

where

$$H_0 = \hbar D(T) S_z^2 + \hbar E_x (S_x^2 - S_y^2)$$

$$H' = g \mu_B B_z S_z + d_z \Pi_z S_z^2$$

For H_0 term, $D(T)$ is zero field splitting which is temperature dependent, since basal defect has the lower symmetry, it has a transverse strain term E_x . and $S_{x,y,z}$ is the electronic spin operator. The eigenstates of the $H_0|\pm\rangle = \frac{1}{2}(|\uparrow\rangle \pm |\downarrow\rangle)$, and $|0\rangle$, which

are the eigenvalues $D \pm E_x$, 0. For H' term, the electron g factor $g = 2.00$, μ_B is the Bohr magneton, B_z is the external magnetic field fluctuation. We also include the magnetic field fluctuation which is represented by longitudinal component B_z . In addition, electric fluctuation can be marked as Π_z [158-159].

In the ODMR measurement by applying an external microwave field with frequency ω that is exactly on resonant the transition. In basis of $(\uparrow, 0, \downarrow)$, the initial Hamiltonian $H = H_0 + H'$ can be expressed as:

$$H = h \begin{bmatrix} D + \Pi'_z + B'_z & 0 & E_x \\ 0 & 0 & 0 \\ E_x & 0 & D + \Pi'_z - B'_z \end{bmatrix}$$

If another external microwave with the frequency ω which is on resonant with $|0\rangle \leftrightarrow |+\rangle$. Then, we need to change to the basis to $(+, 0, -)$, the total Hamiltonian can be written as

$$H_{dr,\pm} = h \begin{bmatrix} D + \Pi'_z + E_x & \Omega \cos \omega t & B'_z \\ \Omega \cos \omega t & 0 & 0 \\ B'_z & 0 & D + \Pi'_z + E_x \end{bmatrix}$$

where Ω is the Rabi frequency, and $B'_z = \frac{g\mu_B}{h} B_z$, $\Pi'_z = \frac{d_z}{h} \Pi_z$. Only the transition between $|0\rangle$ and $|+\rangle$ would be driven, and the transition $|-\rangle$ is decoupled. In the rotating frame with the operator $V = \exp(i\omega t S_z^2)$, and the detuning $\Delta = D + E_x - \omega$. The new Hamiltonian in the rotating frame is

$$H_{rot,\pm} = h \begin{bmatrix} \Delta + \Pi'_z & \Omega & B'_z \\ \Omega & 0 & 0 \\ B'_z & 0 & \Delta + \Pi'_z - 2E_x \end{bmatrix}$$

Going back to the basis $(\uparrow, 0, \downarrow)$, the total Hamiltonian is

$$H_{rot} = h \begin{bmatrix} \Delta + \Pi'_z - E_x + B'_z & \frac{1}{\sqrt{2}}\Omega & E_x \\ \frac{1}{\sqrt{2}}\Omega & 0 & \frac{1}{\sqrt{2}}\Omega \\ E_x & \frac{1}{\sqrt{2}}\Omega & \Delta + \Pi'_z - E_x - B'_z \end{bmatrix} \quad (7)$$

The detuning is $\Delta = D(T) + E_x - \omega$, $B'_z = \frac{g\mu_B}{h} B_z$, $\Pi'_z = \frac{d_z \Pi_z}{h}$. The interesting term in the H is the off-diagonal term E_x , which applies the transition between $|\uparrow\rangle$ and $|\downarrow\rangle$. It would lead to an average effect for the magnetic field fluctuation acting on $|\uparrow\rangle, |\downarrow\rangle$ which is like the dynamical decoupling.

To demonstrate the thermometry, we use a Ramsey fringes scheme. Firstly, we initialize the spin into the state $|0\rangle$, and a $\pi/2$ pulse is applied to drive the system into the superposition of $|0\rangle$ and $|+\rangle$ state with equal amplitude:

$$U_{\pi/2}|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle - i|+\rangle). \text{ Then the system will evolve a time } \tau, \text{ we then use another } \pi/2$$

pulse to read out the phase information to the population of the state. The pulse of the microwave pulse can be on or off by selecting Rabi frequency Ω or 0. When the microwave is off, the Hamiltonian is:

$$H_{rot}(0) = h \begin{bmatrix} \Delta + \Pi'_z - E_x + B'_z & 0 & E_x \\ 0 & 0 & 0 \\ E_x & 0 & \Delta + \Pi'_z - E_x - B'_z \end{bmatrix}$$

Δ, B'_z, Π'_z can be ignored during the operations. When the microwave is on, the Hamiltonian in basis $(\uparrow, 0, \downarrow)$ can be written as:

$$H_{rot}(\Omega) = h \begin{bmatrix} -E_x & \frac{1}{\sqrt{2}}\Omega & E_x \\ \frac{1}{\sqrt{2}}\Omega & 0 & \frac{1}{\sqrt{2}}\Omega \\ E_x & \frac{1}{\sqrt{2}}\Omega & -E_x \end{bmatrix}.$$

If we consider an initial state $|0\rangle$, first a $\pi/2$ pulse is applied $U_{\pi/2} = e^{-i\frac{\pi}{4\Omega}H_{rot}(\Omega)/\hbar}$ that turn the system to superposition of $|0\rangle$ and $|+\rangle$ state:

$$U_{\pi/2}|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle - i|+\rangle).$$

Then, the system will evolve for a time constant τ $U_{\pi/2} = e^{-iH_{rot}(0)\tau/\hbar}$. Then we get

$$U_{\tau}U_{\pi/2}|0\rangle = \frac{1}{\sqrt{2}}|0\rangle + \frac{e^{-2\pi i(\Delta + \Pi'_z - E_x)\tau}}{2i} \left[\left(\cos 2\pi\delta\tau - \frac{E_x + B'_z}{\delta} i \sin 2\pi\delta\tau \right) |\uparrow\rangle + \left(\cos 2\pi\delta\tau - \frac{E_x - B'_z}{\delta} i \sin 2\pi\delta\tau \right) |\downarrow\rangle \right]$$

where, $\delta = \sqrt{E_x^2 + B_z'^2} \approx E_x(1 + \frac{B_z'^2}{2E_x^2})$. Finally, another $\pi/2$ pulse was applied for the

projection and read out of the spin state. The final population of state $|0\rangle$ can be

expressed by $A_{0,final} = \frac{1}{2} - \frac{1}{2}i(A'_\uparrow + A'_\downarrow)$, where $A'_\uparrow(A'_\downarrow)$ is the amplitude of $|\uparrow\rangle(|\downarrow\rangle)$ before the $\pi/2$ rotation. If we neglect all the terms smaller than $O(B'_z/E_x)$, it becomes:

$$A_{0,final} = \frac{1}{2} \left(1 - i e^{-2\pi i \left(\Delta + \Pi'_z + \frac{B_z'^2}{2E_x} \right) \tau} \right)$$

and the final population of $|0\rangle$ is:

$$P'_{0,final} = \frac{1}{2} - \frac{1}{2} \cos 2\pi \left(\Delta + \Pi'_z + \frac{B_z'^2}{2E_x} \right) \tau$$

One should note that the oscillation of the population of P'_0 is decided by the detuning $\Delta = D(T) + E_x - \omega$. Since $D(T)$ will vary with respect to the temperature, and at the same time the environmental noise has neglectable effect on the oscillation frequency. So the frequency of the oscillation P'_0 is sensitive to the temperature and therefore enables the thermometry.

The sensitivity for the thermometry is determined by the spin's coherence time. The magnetic field fluctuation would mostly affect the coherence time. When the spins undergo the evolution, the unexpected B_z has opposite effects on spin $\uparrow$ and $\downarrow$ by and will result in a positive or negative phase for $|\uparrow\rangle$ or $|\downarrow\rangle$ state. Transverse strain will result in a flip between $|\uparrow\rangle$ and $|\downarrow\rangle$ continuously. Therefore, the direction of the magnetic field experienced by the electron spin is changing continuously. If the strain

is big enough, the effect of B_z disappears. Then the environment noise will decrease. Which enables a self-protected mechanism even without the sophisticated dynamic decoupling scheme.

5.3 Realization of the thermometry based on defect spins

The sample we use is the commercially available high-quality semi-insulating (HPSI) 4H-SiC from CREE. We built a confocal microscopy system with microwave feed in to control the spin as displayed in Figure 5-1a and 5-1b. The excitation laser is a 950 nm laser diode. The laser beam is focused on the sample by 0.8 N.A. infrared objective. We estimate that the area that has been excited is around a few microns. The emission was guide to an infrared photo diode after passing the 1000nm long pass filter. The microwave was guided to the sample surface with 80 μm inner diameter ring shape (Au/Pt) antenna fabricated by e-beam. Considering our signal is relatively very small. The microwave field we use is modulated with 20Hz a lock-in technique has been applied as shown in Figure 5-1b.

Figure 5-1: (a) Home-built confocal microscopy system for the thermometry. (a) . The excitation laser is a 950 nm laser diamond. The laser beam is forced on the sample by 0.8 N.A. infrared objective. The emission is guided to the photo detector (Femto, OE-200-IN1). To change the temperature, the sample is mounted in the cryostatation (4K-

300K). We measure the room temperature case in another set up in which the sample was put on a resistive heater (Thorlabs, TEC3-2.5) with a sensor (Thorlabs, HT10K) to measure the temperature. (b) The microwave system we use. The microwave will pass through two switches for two different modulation. First one is the 20Hz modulation for the lock-in technique and the second one is the microwave pulse to control the electron spin of the defects. The photodetector was connected to a lock in amplifier (Stanford Research System SR830) for lock in detection.

5.3.1 Temperature dependence of ODMR spectrum and ZFS (zero field splitting) of PL5 in silicon carbide

Firstly, we measure the ODMR spectrum of the different divacancy defects in silicon carbide. As displayed in Figure 5-2a lower panel, different microwave frequency resonance is clear in the ODMR spectrum without any magnetic field applied. To figure out where the peaks come from, we vary the magnetic field strength in the z direction and measured the ODMR which shows in Figure 5-2a upper panel. Different types of divacancy have different symmetry which result in different diverging behavior with respect to the magnetic field. For instance, when the magnetic field is zero, there is very small splitting between the two transitions. If the magnetic field is added, the two transitions will split to left and right with the same slope 2.8 MHz/G [15] as marked using dash line in the figure. This ODMR resonance with respect to the magnetic field is very similar to that of the NV center. We identify these defects as PL6. While PL5 has the C_{1h} symmetry, unlike PL6, two resonance transitions will only bend. We pick up the right branch ODMR resonance of PL5 because it is not mixed with other resonance like PL7's.

We also measured the ODMR resonance with respect to different temperatures. The ODMR resonance frequencies will increase if the temperature goes down [160]. This phenomenon is because of the thermal expansion as well as the interaction between

electron and phonons. As displayed in the ODMR resonance scan shown in Figure 5-2b, a PL5-PL7 show the same behavior, which implies that such shift of the resonance is their intrinsic property. The zero field splitting of the PL5 with respect to the temperature from 20K to 300K is in Figure 5-2c and this shift can be fitted with a fifth-order polynomial. From the left resonance ($D - E_x$) and right resonance ($D + E_x$), the transverse strain is estimated to be E_x (16.5 ± 0.048 MHz). The temperature dependence of the E_x has also been shown in the figure and the slope is around -0.7 Hz/K, it implies this term $2E_x$ is not sensitive to the temperature[159]. The shift of the ZFS of the PL5 are mainly due to the D temperature dependence, not E_x . To get the detailed relationship between D and the temperature, we got the ODMR frequency of PL5 right peak ($D + E_x$) around the room temperature as displayed in Figure 5-2d. We found that D follows a good linear relationship with a slope of $dD/dT = -109.4 \pm 0.4$ kHz/K. It shows that the OMDR resonance in PL5 is more sensitive to the temperature, in comparison with those in NV centers ($dD/dT \sim -74.2$ kHz/K).

Figure 5-2 (a) ODMR resonance map of different divacancy in 4H-SiC. Different types of divacancy has been marked as PL5, PL6, PL7. (b) Temperature dependence of ODMR resonance. Dashed red line is the center of the resonance. Lower panel shows the ODMR resonance at 300K. (c) Upper panel: Temperature dependence of ZFS (Zero Filed Splitting) of the ODMR spectrum. Lower panel: Transverse strain with respect to the temperature, shows that it is not sensitive to the temperature change. (d) ZFS close room temperature. A linear relationship between the ZFS and the temperature is obvious.

5.3.2 Coherent control of the PL5 divacancy spins in silicon carbide

The linear relationship between ZFS with respect to the temperature shows that it can be potentially used in the thermometry by Ramsey fringes thermometry. We successfully demonstrate coherent control the divacancy spins PL5 in SiC as shown in Figure 5-3. Figure 5-3a shows the pulse sequence we use including Rabi oscillations and Ramsey fringes. At first, a $\frac{\pi}{2}$ -pulse is applied to the system to initialize the spin state, and then either a Rabi or Ramsey pulse is added with 20 Hz lock in modulation. Finally, another 2 μ s pulse is applied to read out the spin states. The fluorescence change

ΔPL will be detected with lock-in method. Figure 5-3(b) and Figure 5-3(c) displays Rabi oscillations and Ramsey fringes at the temperature is 210K. The Ramsey fringes is fitted with the equation

$$I = a \exp \left[-\left(\frac{t}{T_2^*} \right)^n \right] \cos(2\pi f t + \phi) + b$$

In which b , a , ϕ and n are free parameters, t and T_2^* are the evolution time and dephasing time respectively. Please note that the microwave was intentionally detuned by 2MHz from the resonance and thus induce the oscillation of the Ramsey fringes. The dephasing time T_2^* is a very important parameter in the defect-based sensing. We measure Ramsey fringes at different temperature and from the fitting we extracted T_2^* as shown in Figure 5-3(d). From our measurement, T_2^* remains is not sensitive to the temperature and remain 2.2 μs from 20K to 300K.

Figure 5-3 (a) Pulse sequence for Rabi oscillations and Ramsey fringes and Ramsey pulse sequence. A $\frac{\pi}{2}$ -pulse is applied to the system to initialize the spin state, and then either a Rabi or Ramsey pulse is added with 20 Hz lock in modulation. Finally, another 2 μs pulse is applied to read out the spin states. (b) Rabi oscillation of PL5 divacancy spins in silicon carbide. Intensity change has been plotted as a function of microwave pulse duration. The red balls are the raw data and blue solid line is the fitting. This is measured at the temperature of 210K (c) Ramsey fringe of PL5 divacancy spins in silicon carbide. the microwave was intentionally detuned by 2MHz from the resonance and thus induce the oscillation of the Ramsey fringes. From the fitting we extract the dephasing time T_2^* is 2.2 μs. (d) T_2^* time with respect to temperature. We conclude that the dephasing time is not sensitive to the temperature.

Figure 5-4 (a) Ramsey measurement at 293.3K. The red dots are fitted with Equation (14). (b) The oscillation frequency of Ramsey fringes $\Delta(T) = D(T) + E_x - \omega$ as a function of temperature while the microwave frequency ω remains constant. Frequency is fitted to be linear with the temperature with a slope of $-108.2 \pm 0.5 \text{ kHz/K}$.

5.3.3 Ramsey fringes-based thermometry

We perform thermometry with the Ramsey fringes method. The frequency of the oscillation of the Ramsey fringes is intentionally introduced by detuning Δ . As a result, the temperature change will lead to a linear change of D value as displayed in Figure 5-3(d). Figure 5-4(a) shows that Ramsey measurement of PL5 has pure oscillations without interaction with nuclear spins. Figure 4(b) shows the oscillations of the Ramsey oscillation frequency has a very good linear relationship with the temperature change with a slope of $-108.2 \pm 0.5 \text{ kHz/K}$. Note that this slope agrees well with the measured temperature dependence shift $-109.4 \pm 0.4 \text{ kHz/K}$ of ODMR resonance spectrum. The thermometry based on Ramsey fringes can be applied to a big range of temperature, as displayed in Figure 5-2(c). We estimate the sensitivity η to be about $205.6 \text{ mK/Hz}^{1/2}$ by

the method mentioned in.

5.3.4 Thermo echo-based thermometry

Another most frequently used schemes of the thermometry are the Thermal Echo (TE) sequence. In the Thermal Echo, the middle microwave pulse was replaced by a 2π pulse when the spin is evolving in the superposition state of $|\uparrow\rangle$ and $|\downarrow\rangle$. With this pulse, the two state exchange their population and cancel the asymmetric phase accumulations $B'_z S_z$.

In the case of NV center in diamond, if the system is used a TE sequence, T_2^* can be increased at least an order of magnitude as those in the Ramsey fringes method. In the PL5 divacancy case, there is not this expected enhance effect because B'_z has already been suppressed for Ramsey fringes measurement.

We then apply the TE pulse sequence to demonstrate thermometry. As displayed in the Figure 5-5(a), the coherence time extracted from the fitting is $2.3\ \mu\text{s}$ which is very close to the dephasing time in the Ramsey experiments. This can be reconfirmed with our theoretical model. Again Figure 5-5b shows that the frequency has a good linear relationship with the change of temperature. The slope $-108 \pm 1\ \text{kHz/K}$ agrees with the temperature dependence of the D value very well.

Figure 5-5: (a) TE sequence at room temperature. (b) The frequency extracted from TE sequence vs temperature.

As discussed above, the PL5 have a large transverse strain E_x , and the transverse strain term will act as a protector to the system to the environmental noise. We verify this mechanism by the following experiments. First, we introduce a magnetic field noise which has an amplitude of B (so the magnetic field will vary from $-B$ to B at most). The frequency of the change is 100 Hz. At the same time, we increase B and measure Ramsey fringes to observe how the dephasing time will react as the change of B . The result is as Figure 5-6, we extracted T_2^* for different strength of magnetic field noise case. The simulation is done by the Hamiltonian as in previous sections. When we do the experiment, the applied magnetic field is in the z direction and in that case B_z' in Equation (13) equals to $B_z' = \cos(109.5^\circ)B_{\text{applied}}$, where B_{applied} is total amplitude of the B field. Two free parameters in Equation (13), namely Π_z' and B_z' . For Π_z' , we tentatively assume it has the standard normal distribution with zeros the be mean value. The standard deviation $\sigma_{\Pi_z'}$ is determined by the experimental result $T_{2,B_z=0}^* = 1.8 \mu s$ when the magnetic frequency is 0 MHz.

Figure 5-6 shows that the red dots theoretical simulation agrees with the experimental data quite well. When $E_x = 0$, T_2^* decreases dramatically with respect to the strength of the magnetic field strength. It agrees with the previous experimental results which is

done in NV center in diamond that the dephasing time T_2^* is very sensitive to the magnetic field noise. Till now, our theoretical models and the simulations both support that the non-zero transverse strain term E triggers a self-protected mechanism of the spin with respect to the magnetic field noise and also ensure the excellent sensitivity of the spin-based thermometry.

Figure 5-6 Dephasing time T_2^* and magnetic noise strength (black dot for experiment data; red and blue dot for simulation). For the PL5 divacancy spin with transverse strain term $E=16.5$ MHz, T_2^* decays much slower than the case when $E=0$, which confirms the self-protected mechanism with E_x in SiC PL5 defects.

5.4 Conclusions and outlook

To conclude, we have performed the divacancy spin-based thermometry based on the Ramsey fringes and thermal Echo respectively. Our experimental agrees well with the simulation. More importantly, we found the large transverse strain will induce a

self-protection effect to the system to environmental magnetic noise. By employing more sophisticated scheme such as dynamical decoupling[161], or to get purification of the SiC[162], and also the methods to further increase our collection, like solid immersion lenses[84], nanopillars [86], the sensitivity of the thermometry will be further increased.

One should note that the silicon carbide itself is bio-friendly and it will cause less damage to the cells if infrared photons are used. Thus, it is easy to think that this technique is valuable to the telemetry in a living cell as is already demonstrated in the NV center in diamond case. Moreover, if the SiC nanoparticles [88, 163] and the atomic force microscope (AFM) are combined, it can be used in the nanoscale thermal image[164].

Chapter 6 Conclusions and Future Work

6.1 Conclusions

To conclude, even many SPSs have been reported, searching for new SPSs that has better optical properties is still a hot topic. I have presented our discovery of telecom wavelength SPSs in both gallium nitride and silicon carbide. They are bright with MHz repetition rate and high single photon purity. More importantly, they have a relatively high degree of polarization (93.2% for SPSs in GaN and 97% for SPS in SiC) which is useful in the QKDs. The mature fabrication technique, the familiarity of the whole industry will favour the on-chip integration of these newly founded SPSs.

To achieve a quantum network in the long run, how to scalable and deterministically fabricate those SPSs is the central technique. I also presented our work on direct writing of single germanium arrays in diamond based on focus ion beam technique. The single photon generation ratio can reach 53.33% at the dose of 200Ge^+ . It means if we implanted 200Ge^+ at a specific position in the diamond, there is more than 50% possibility that we can get a single germanium vacancy center. This technique will be very helpful if one wants to couple the germanium spin with photonic structures like nanocavities or waveguides.

The spin control is another his work shows that the all-optical control the spins in the transition of SiV is possible. central technique in the area, all optical control is necessary for both underlying the physics behind the energy levels and for simplifying the spin control protocol. I presented our work on coherent control of a strongly driven optical transition in silicon vacancy in diamond. The time domain optical Rabi

oscillations and Mollow triplet spectrum have been demonstrated.

For real quantum applications, the spins in the solid have been explored to be sensors. Because of their atomic scale and high sensitivities to the outside field, those quantum sensors usually behave remarkably. Finally, I presented our work that demonstrates an example of quantum application: nanoscale temperature sensing based on the divacancy spins in silicon carbide. The spatial resolution can reach hundreds of nanometers (the size of the excitation laser beam by the diffraction limit). The sensitivity of the thermometry can reach up to $200 \text{ mK/Hz}^{1/2}$. Our method is extremely useful in the microelectronics and biological applications.

6.2 Future possible directions

6.2.1 Two-photon interference of GeV center in diamond

Generation of indistinguishable photons is one of the central tasks in many quantum applications. The coherent photons with excellent optical properties have been demonstrated in silicon vacancy in diamond in [165]. The reason is that because of the inversion symmetry of the SiV, the energy level is not easy to be affected by the environment field which results in the lifetime-limited linewidth founded in silicon vacancy in diamond. Also, the strain effect in the bulk diamond is not strong so that it is possible to find two neighboring SiVs that have almost the same resonant optical frequencies.

Figure 6-1 Schematic of set up for the two-photon interference between two nearby SiVs. a. Two scanning mirrors correspond to two integrated confocal microscopy system that each one excites on single SiV individually and ZPL and PSB part are collected respectively. b. The ZPLs from two SiVs will be guided to a same beam splitter to form an Hong-Ou-Mandel (HOM) interference set up. c. The emission spectrum before and after etalon, the etalon is used to filter our one peak individually. The reason is SiV has the characteristic ABCD structures that have hundreds of GHz splitting, to achieve interference, there must be only one peak been filtered out [165].

As shown in Figure 6-1, to demonstrate the two-photon interference, two confocal microscopy system needs to be integrated together in order to excite and collect the emission from nearby SiVs. Two SiVs are excited individually and their ZPL and PSB are collected respectively. To get the interference pattern, a Hong-Ou-Mandel set up as Figure 6-1c is needed. Two zero-phonon lines from two nearby SiVs will interfere in the beam splitter.

Figure 6-2 HOM interference experiment between two nearby SiVs. The interference pattern of the correlation between the two ZPLs from nearby SiVs, same polarizations for the pink color and different for the green color. As expected, $g^2(0)$ for pink is far less than green [165].

The results are shown in Figure 6-2. It is a correlation measurement between two ZPLs from nearby spins. The polarization of the two ZPL can be adjusted to be same or different, if the polarization is the same, then those emission from two SiVs are closer to the indistinguishable photons and the $g^2(0)$ will be closer to 0 than whose polarization is different.

For germanium vacancy center, it has similar optical properties as silicon vacancy center in diamond, such as same spin-orbit coupling induced ABCD transitions, same inversion symmetry to protect the optical transitions undergo different mechanism of broadening. So, it is highly possible that the two-photon interference can be also achieved in the germanium vacancy system.

6.2.2 Nano-fabrications with SPSs

Researchers are exploring more ways to engineer SPS' optical properties. One of the methods is to fabricate special photon structures like pillars, waveguides or photonic

cavities.

Figure 6-3 The fabrication process of pillars in silicon carbide [86].

6.2.2.1 Pillar structures

One structure is the pillar as Figure 6-3 shows, if the emitter is sitting in the middle of the pillar, because of the Purcell effect, the enhancement of the spontaneous emission will be achieved [86]. This is a way to further increase the SPS's emission rate.

6.2.2.2 Chirped diamond grating

Figure 6-2 **a**, The diamond-air interface, because of the high refractive contrast between Air and the diamond. The angle that total internal reflection is small leading to great loss of the total emission. **b**, The chirped diamond grating to enhance the emission of the grating, by fabricating this structure, the near-unity collection efficiency of the NV center has been achieved. **c** Simulation from the side, there is two interfaces, air to the grating and grating with the diamond [166].

Another possible structure is the chirped diamond grating which is fabricated on the diamond membrane [166]. It is a designed photonic structure and the emitter is sitting in the center of the grating. As Figure 6-2a shows that because of the high refraction index between diamond and the air, the loss of the emission is huge. Only a small fraction will be finally collected to the system. Researcher designed a chirped diamond grating on top of the NV center and based on the simulation and the results, they claimed that the collection efficiency of the whole system can reach to unity which means by the help of this structure, almost all the photons that emit from the single NV center has been collected without any loss.

6.2.2.3 Integration with photonic waveguides

Figure 6-3 On-chip excitation of single germanium vacancies with photonic waveguides. The nanodiamond containing is embedded in the center of the waveguide. The 532 nm laser was excited on the left, coupled into the waveguide with the help of the grating, the laser field propagates along the waveguide and the GeV embedded in the center will be excited, the emission will travel along the waveguides, emit to the air through the grating [167].

The on-chip excitation of single germanium has been achieved in [167]. The two sides of the waveguide are the grating structures which is used to free space-waveguide coupling or waveguide-free space radiation. As shown in the Figure 6-4, the

nanodiamond containing single GeV center has been embedded in the center of the waveguide. The excitation was done from the left of the waveguide, the 532nm was coupled from the free space to the waveguide through the grating. When the laser field propagates along the waveguide, it will excite the GeV center in nano diamond. The emission will also travel along the waveguides and collected on the right with the help of the other grating. The on-chip excitation of the single GeV has been achieved based on the scheme.

6.2.3 Spin photon entanglement with solid state spins

As I mentioned above, the proposed quantum internet consists of two parts: the stationary qubits for the quantum information processing and storage [39]. So, the realization of the entanglement between stationary qubit and the flying photons are in the central importance. Spin photon entanglements have been demonstrated in NV centers [117] and quantum dots [114] recently.

The main entanglement is as shown in Figure 6-4, it shows two types of Λ system that can be used in the entanglement protocol. The photons have two main freedom: frequency and polarization. Both can be coupled with the electron spin.

In the left picture, the electron spin can be entangled with the colour and polarization. When the system emits the red photon, it means that it is in the spin-up state. On the contrary, when it emits the blue photon, it is in the spin-down state. For the right figure, the energy level gap is the same, but the polarization of the emitted photons is opposite. By detecting the polarization of the emitted photon, the spin state can be read out.

Figure 6-4 Λ system in spin photon entanglement [168].

In some system like germanium vacancy centre in diamond spin defects in silicon carbide, these Λ system can be also found and it is possible to realize spin-photon entanglement in these spins.

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