

Investigation of the silicon vacancy color center for quantum key distribution

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Abstract: Single photon sources (SPS) are crucial for quantum key distribution. Here we demonstrate a stable triggered SPS at 738 nm with linewidth less than 5 nm at room temperature based on a negatively charged single silicon vacancy color center. Thanks to the short photon duration of about 1.3-1.7 ns, by using high repetition pulsed excitation at 30 MHz, the triggered single photon source generates 16.6 kcounts/s. And we discuss the feasibility of this triggered SPS in the application of quantum key distribution.

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1. Introduction

Demonstrations of quantum key distribution (QKD) have been achieved with a variety of photon sources, such as attenuated laser pulses, photon pairs from parametric down conversion, and single photons from quantum dots or individual color centers in diamond [1–7]. Among them, true single photons generated by single emitters can demonstrate high reliability when compared to an attenuated pulsed laser [4]. Quantum dots can emit broadband single photons covering the telecom band, but have drawbacks including cryogenic temperature operation and fluorescence intensity instability. Diamond color centers are usually stable at room temperature, and thus are ideal candidates for a true SPS. Many kinds of color centers in diamond, emitting at different wavelengths, are possible dependent upon the elemental content and structure of the impurity. So far, nitrogen vacancy color

centers (NV), silicon vacancy color centers (SiV), nickel-related color centers, and chromium-related color centers have been considered as candidate SPSs [8–14].

The NV center has been studied for implementation in QKD [4], however the NV center has a broad photoluminescence spectrum covering 600 – 800 nm due to phonon broadening, which prevents efficient filtering of signal photons from background light. In addition the NV excited state lifetime is about 10 – 20 ns, with NV centers in nanodiamond exhibiting longer lifetime than their counterparts in bulk diamond. In combination with the presence of metastable shelving states, the long fluorescence lifetime means fluorescence count rates above 10^6 are difficult to achieve [15–17]. As the NV emission is reported to occur via two orthogonal dipoles, the polarization contrast of a single NV center is usually sub-unity [18]. These properties limit the application of NV centers to QKD.

It was reported recently that the negatively charged silicon vacancy color center (SiV⁻) has narrow emission around a zero phonon line (ZPL) of 738 nm with a short excited state lifetime, usually about 1 ns [19–26]. The polarization contrast of the SiV⁻ can reach almost 100% when viewed from the correct plane [21]. Therefore the SiV⁻ center could be used to improve the transmission speed as well as the signal to noise ratio in free-space QKD. In this paper, we present a triggered SPS by employing single SiV⁻ centers in a bulk diamond at ambient temperature. Under pulsed excitation with our scanning confocal microscope setup, we observe SiV⁻ centers exhibiting a ZPL at 738 nm with narrow linewidth of 5 nm full-width at half-maximum (FWHM), short photon duration of about 1.3-1.7 ns, as well as 50% linear polarization contrast at room temperature. Due to the short photon emission lifetime, high repetition excitation could be implemented. With an excitation repetition rate of 30 MHz, the maximum recorded photon count rate was 16.6 kcounts/s. Our experimental results indicate that pulsed laser excitation of SiV⁻ has great potential as a stable triggered SPS with applications in higher key rate and improved safety-level quantum key distribution.

2. Sample preparation and experimental setup

The SiV⁻ color centers used in the experiment were fabricated by low-energy implantation of silicon atoms into a bulk diamond substrate [27,28]. First, a thin homoepitaxial diamond film with thickness of 27 μm was grown on (100)-oriented type-Ib HPHT diamond by microwave plasma chemical vapor deposition. Doubly charged silicon ions were implanted into the homoepitaxial diamond surface at an acceleration energy of 60 keV by using a focused ion beam system. Thermal annealing for the formation of SiV⁻ centers was performed at 1000 °C in 10% H₂ forming gas for 30 min. With this method, bright SiV⁻ centers were fabricated with spatial control allowing for investigation of individual defects.

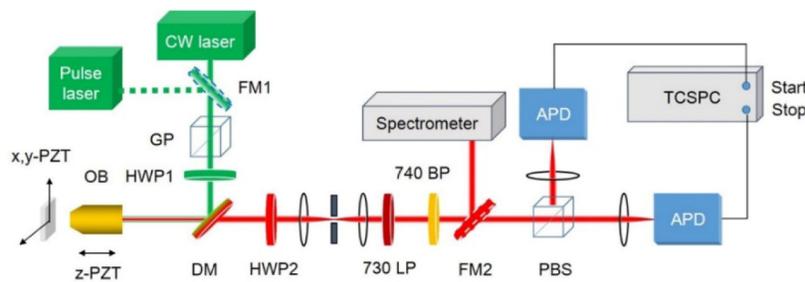


Fig. 1. Scheme of the experimental setup. FM1, 2, flip mirrors. GP, Glan prism. HWP1, 2, half-wave plates. DM, dichroic mirror reflecting excitation laser of 532 nm and passing the fluorescence longer than 540 nm. OB, objective. PZT, piezoelectric ceramic transducer. 730 LP, long-pass filter cutting off at 730 nm. 740 BP, band-pass filter, bandwidth ~10 nm. PBS, polarized beam-splitter. APDs, single-photon counting modules based on avalanche photodiode. TCSPC, time-correlated single-photon counter.

The experimental setup is a homemade confocal microscope, shown in Fig. 1. The diamond sample was placed on an x-y stage and translated with a piezoelectric ceramic transducer (PZT). The focus point along the z-axis was adjusted with a PZT on the microscope objective. The excitation laser source was either a continuous-wave (CW) solid-state green laser at 532 nm, or a pulsed laser at 532 nm with 30 MHz repetition rate and 15 ps pulse duration. The laser source was focused on the diamond surface with an oil-immersion microscope objective (Olympus, N.A. = 1.40, 100 \times) and the fluorescence from the color center was collected by the same microscope objective. After spatial and spectral filtering, the fluorescence was sent to the detection part of the setup which consisted of a polarized beam splitter (PBS) with two single-photon detectors (Perkin Elmer, SPCM-AQR-14) at each output port in Hanbury-Brown and Twiss configuration. A half-wave plate (HWP2 in Fig. 1) could be used to adjust the splitting ratio of the PBS to balance the intensity of light incident on the APDs at the two ports. The outputs of the single-photon detectors were connected to a time-correlated single-photon counter (TCSPC, Pico Harp 300, PicoQuant GmbH). Fluorescence from the sample could also be sent to a spectrometer for spectral analysis by a flip mirror.

3. Experimental result

A fluorescence image of an investigated single SiV⁻ center is presented in Fig. 2(a). The diameter of the fluorescent spot is about 300 nm as shown in Fig. 2(b), which is close to the diffraction limit of the microscope objective. The photoluminescence spectrum of the SiV⁻ center is shown in Fig. 2(c), and the fluorescence wavelengths selected for investigation, by a 740 nm bandpass filter with bandwidth of 10 nm, marked by the orange shaded area. The collected ZPL emission of the SiV⁻ center showed a linear polarization along $\{110\}$ direction with polarization contrast of about 50% as shown in Fig. 2(d), which agree with the expected SiV⁻ color center fluorescence polarization for this diamond orientation [19]. The polarization contrast here didn't achieve 100% because both of excited state and ground state of a SiV⁻ have a doublet fine structure and there are four transitions of different linear polarization directions. We believe the 50% polarization contrast is a mixed result of the four transitions when the SiV⁻ was viewed from (100) plane.

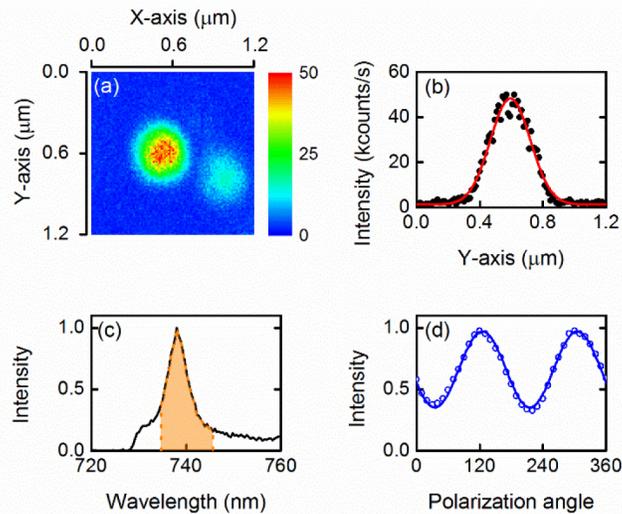


Fig. 2. (a) Fluorescence scan image of the sample under CW excitation; (b) Linescan across the SiV⁻ center along Y-axis of (a); (c) Spectrum of the SiV⁻; (d) Polarization property of the emitter.

The unitary of the SPS was evaluated by a photon-correlation measurement with the HBT setup. Measured photon coincidences $c(\tau)$ during measurement duration T with measurement resolution ω under CW laser excitation can be normalized to the auto-correlation function by

$$C_N(\tau) = \frac{c(\tau)}{N_1 N_2 \omega T}, \quad (1)$$

where N_1 and N_2 are the counting rates on the two single-photon detectors. With background correction, the photon correlation function can be written as

$$g_c^2(\tau) = \frac{C_N(\tau) - (1 - \rho^2)}{\rho^2}, \quad (2)$$

where $\rho = S/(S + B)$, and S represents the signal counting rate and B the background counting rate. For an ideal SPS, the coincidence at zero time delay $g_c^2(0)$ equals zero. Therefore, evaluation of a practical SPS depends on its $g_c^2(0)$ value. As shown in Fig. 3(a), when the CW excitation power was 6.9 mW, the two detectors recorded 53 and 50 kcounts/s, respectively. Using a TCSPC with a temporal resolution of 0.128 ns and integration time of 300 s, a value of 0.32 is expected for $g_c^2(0)$ according to Eqs. (1) and (2), for a single emitter. However, due to the instrumental response function (IRF) time of about 660 ps, $g_c^2(0)$ was increased slightly since under CW excitation there exists a certain possibility that the single SiV⁻ emitted a second photon within the system IRF time. Since the two successive single photons were very close at time-scale and were separated by the PBS in the HBT, the TCSPC would regard the arrival time of the two photons as the same, leading to the unexpected increase of $g_c^2(0)$. Photon bunching was also observed for an excitation power close to saturation. We note that the bunching decayed in a few nanoseconds providing evidence of a three-level system including a “shelving state” with few nanosecond lifetime [26]. The population dynamics of the three states in such a system was discussed in Ref [10], where nickel-related color center was studied. According to such a model, fast quench of the bunching effect in the photon-correlation measurement means the lifetime of a shelving state is short. Therefore, a big photophysical difference between a SiV⁻ and a nickel-related center we can deduce from the decay time of bunching effect is that the “shelving state” or “metastable” state of SiV⁻ has a much shorter lifetime.

Next we switched the excitation source to a pulsed laser in order to avoid experimental issues associated with the finite temporal resolution and IRF of the detectors. The repetition rate was 30 MHz with a pulse duration of 15 ps, much shorter than the emission lifetime of the SiV⁻ center so that a second excitation of the SiV⁻ within a single excitation pulse could be avoided. When the excitation power was 0.99 mW, both detectors recorded 8.3 kcounts/s. Under excitation with pulsed laser, the measured total coincidence counts $c(m)$ in peak m during measurement duration T with pulse repetition period θ , can be normalized to give

$$C_N(m) = \frac{c(m)}{N_1 N_2 \theta T}. \quad (3)$$

With a repetition period θ of 33 ns and measurement time T of 1000 s, we experimentally observed photon-correlation under triggered emission displaying $C_N(0) = 0.095$ as seen in Fig. 3(b). We normalized the peak-area coincidences marked at top of each peak according to Eq. (3), verifying the unitary nature of the emitter. Comparing $C_N(0)$ measured under pulsed excitation to $g_c^2(0)$ under the CW excitation, with the same SiV⁻ center, the value of $C_N(0)$ is much smaller because by using pulsed excitation, the temporal interval between the sequential photons (33 ns) was much longer than the detection system’s IRF time. In addition, excited state lifetime extracted from this measurements was found to be 1.3-1.7 ns.

We also investigated the dependence of $C_M(0)$ and $[S/(S + B)]$, i.e. the signal counts to total photon counts ratio, as a function of excitation power as shown in Fig. 4(a). Each measurement was integrated for 10 minutes and the coincidences were normalized with Eq. (3). With the an increase in excitation power, more noise photons from the excitation laser source transmit through the filters together with increased background fluorescence from the diamond and the immersion oil. These effect a small drop of $S/(S + B)$, from 0.85 to 0.8 at high excitation power, however the overall impact on the value of $C_M(0)$ is nearly negligible, and it remained small even at high powers.

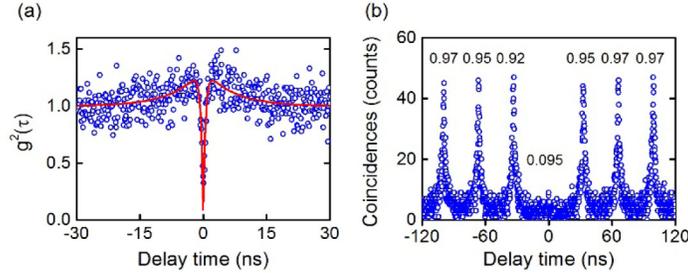


Fig. 3. (a) Photon correlation measurement under CW excitation. Blue circles are normalized values by Eqs. (1) and (2) of measured coincidences. Red curve is a fit by auto-correlation function. (b) SiV fluorescence photon-correlation with pulsed laser excitation. Blue circles are measured coincidence counts.

The brightness of the single emitter was evaluated by measuring the fluorescence intensity I as a function of the pulsed excitation power P , as shown in Fig. 4(b). The experimental data was fitted by the saturation function

$$I = I_{\infty} \times \frac{P}{P + P_{sat}}, \quad (4)$$

where I_{∞} is the maximum limit of fluorescence intensity and P_{sat} is the saturation power. The maximum intensity as fitted was 19 kcounts/s and saturation power as fitted was 0.60 mW. Photon-bunching was not observed under pulsed excitation even when the excitation power was 0.99 mW, far above saturation power, this is because as shown in Fig. 3(a), the photon bunching decayed in a few ns, well before the next excitation pulsed arrived after 33 ns.

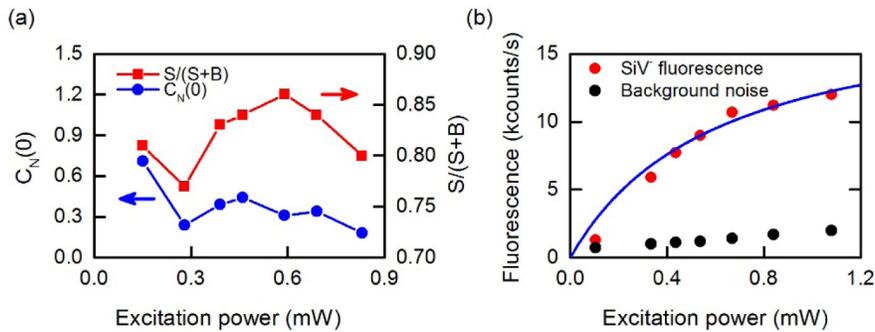


Fig. 4. (a) $C_M(0)$ and $S/(S + B)$ as a function of the excitation power of the pulsed laser. (b) Excitation saturation under pulsed laser excitation. Black dots were background photon counts measured at the same plane of the SiV in the diamond sample.

4. Discussion

The triggered SPS in our experiment showed short photon duration, narrow linewidth and 50% polarization contrast, thus benefitting higher repetition rate and signal to noise ratio, and polarization encoding for QKD. The maximum recorded intensity of the triggered single photon source in this experiment reached 16.6 kcounts/s with an excitation laser repetition of 30 MHz. Since the excited state lifetime is short (1.3-1.7 ns), in principle it should support single-photon generation rates up to hundreds of MHz with pulsed excitation rates of GHz. However, these experiments were limited by the 20-ns deadtime of the single-photon detectors currently employed, therefore limiting the repetition rate of the pulsed excitation source to below 50 MHz in order to avoid missing photodetection events. But, with development of new generation single-photon detectors for QKD, a detector with shorter deadtime would be expected to allow high speed single-photon generation.

Modification of the SiV⁻ sample can be used to raise the collection efficiency and achieve higher photon count rates. By using SiV⁻ in nanodiamonds, fluorescence collection efficiency can be increased [6]. However, drawbacks include a longer excited state lifetime and wider ZPL emission band for SiV⁻ in nanodiamonds [6,14]. Another solution is to use solid-immersion lenses (SIL) which allow the collection efficiency to be enhanced 10 fold without ZPL broadening [16,18]. Surface plasmon resonance enhancement provides another solution for generation of brighter single photon sources [27,28]. Each of these solutions build upon the beneficial intrinsic properties of the SiV⁻ defect, demonstrated here, to enhance the intensity rate of triggered single photon sources.

Chromium-related color centers in diamond are also bright and of similar excitation state lifetime and its quantum yield shifts versus temperature. At cryogenic temperature, it can achieve almost unitary quantum yield, thus have great potential in making great single photon source [29,30]. And recently, a new single photon source in silicon carbide was investigated to be very bright, and is very promising for making excellent single photon source at different band from 648 nm to 677 nm too [31].

5. Conclusion

In conclusion, we achieved a triggered SPS with narrow linewidth, short photon-duration and very low noise using a single SiV⁻ color center under high speed pulsed picosecond laser excitation. The photoluminescence properties of single SiV⁻ promote the value of it for quantum key distribution as the following reasons. Firstly, with careful spectral filtering, the useful narrow spectral emission is easy to be selected from the stray light noise, providing a high signal to noise ratio. Secondly, due to the short photon emission lifetime, high repetition pulsed excitation could be employed to build a fast SPS for high speed QKD. Moreover, narrow time-window analysis on the detection is also compatible with such triggered SPS due to the short photon emission lifetime. Our experimental results promote the value of the SiV⁻ color center for quantum key distribution, and its emergence as a true single-photon source for higher key rate and safety-level photon generation.

Acknowledgments

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