Single photon emission from SiV centres in diamond produced by ion implantation

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Abstract

We report the observation of single photon emission from single SiV (siliconvacancy) centres in diamond produced by ion implantation. The high photostability and the narrow emission bandwidth of about 5 nm at room temperature make SiV centres interesting as a single photon source in practical quantum cryptography. We discuss problems that arise from the nonradiaditve transitions which lower the brightness of the source.

The development of reliable devices for the generation of single photons is crucial for many applications such as, for example, quantum cryptography [1], optical quantum computation [2] as well as experiments on the foundations of quantum optics [3, 4]. Many implementations of single photon sources have been demonstrated [5–8]. One of the most promising candidates is a single photon source based on optically active defects in diamond, the so-called colour centres. The colour centres in diamond are well localized and photostable at room temperature, therefore a simple setup suffices for a single photon source. For single NV (nitrogen-vacancy) centres the generation of single photons has been demonstrated [7], and consequently has already shown advantages compared to attenuated laser pulses in a quantum cryptography experiment [9]. The performance, however, is still not satisfactory for practical applications. The main disadvantages of NV centres as single photon sources are their broad spectrum and relatively long photoluminescence lifetime.

More than 500 optical centres in diamond are documented [10], enough reason to look for more suitable colour centres for single photon generation. Recently, it was shown that single NE8 (nickel-nitrogen complex) centres can emit single photons efficiently with an emission bandwidth of 1.2 nm at room temperature [11]. A further advantage of NE8 might become the fact that they can also be fabricated in CVD (chemical vapour desposition) diamond [12]. Here, we present an alternative way to produce single colour centres in diamond. Ion implantation allows us to deposit any desired impurities into diamond. Impurity density and

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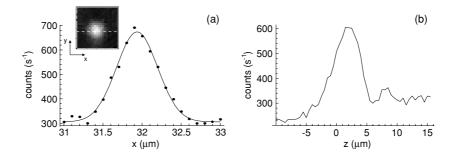


Figure 1. (a) Luminescence intensity profile in transversal direction of a single SiV centre. The inset shows a fluorescence image of this SiV centre as it appears in our confocal microscope. (b) Scan of the SiV centre along the optical axis. The diamond surface was located around $z=0\,\mu\mathrm{m}$.

location can be precisely controlled by the ion dose and ion optics [13, 14]. We employed ion implantation to produce single SiV centres in diamond.

The fluorescence of SiV centres was first observed in 1981 by Vavilov *et al* in cathodoluminescence investigation of polycrystalline CVD diamond [15]. In 1995, this colour centre could be finally unambiguously verified to involve silicon impurities [16]. A theoretical model of a silicon-vacancy complex was proposed by Goss *et al* [17]. The most interesting properties of SiV centres related to the use as a single photon source are their spectrum, which consists of a sharp ZPL (zero phonon line) at 738 nm and only very weak vibronic sidebands at room temperature, and a very short photoluminescence lifetime of 1–4 ns [18].

The implanted samples are IIa diamond windows with dimensions of $0.5 \times 0.5 \times 0.25 \text{ mm}^3$. The top and bottom faces are polished along the (100) plane. One of the main faces was implanted with 10 MeV Si²⁺ ions at room temperature with an ion dose of 10^9 cm^{-2} . The longitudinal range and range straggling of the silicon ions were estimated by a computer simulation with SRIM to be $2.3 \pm 0.16 \,\mu\text{m}$ [19]. Post-implantation annealing was carried out at $1000\,^{\circ}\text{C}$ for 5 min in vacuum.

The photoluminescence measurements were performed by a scanning confocal microscope at room temperature. This system allows us to excite single colour centres in the samples with a spatial resolution of 600 nm in lateral direction and about 3 μ m along the optical axis [7]. The luminescence excitation is provided by a cw laser diode with central wavelength at 685 nm and maximal optical power of 50 mW. In order to generate a proper Gaussian mode, the excitation laser light is coupled into the confocal microscope through a single mode fibre. The fluorescence of the SiV centres is collected into a single mode fibre, too, and can be thus easily guided to a homemade grating spectrometer for spectral analyses, or a Hanbury–Brown–Twiss setup for the measurement of the autocorrelation function $g^{(2)}(\tau)$. To detect low luminescence intensities from single quantum transitions, silicon avalanche photodiodes with a dark count rate of about 200 s⁻¹ and a deadtime near 1 μ s were used for photodetection. The overall detection efficiency of our setup was estimated to be 1.5×10^{-4} .

The inset in figure 1(a) shows a fluorescence image of a single SiV centre behind an interference filter with a central transmission wavelength at 740 nm and a bandwidth of 10 nm. The profile of the luminescence intensity along the dotted line on the fluorescence image is shown in figure 1(a). The transverse width of 622 nm corresponds to the resolution of the confocal microscope. The central peak in figure 1(b) shows the position of the SiV centre in the direction perpendicular to the implanted diamond surface. Outside the crystal (left of the

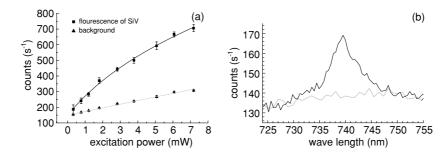


Figure 2. (a) Saturation of the fluorescence of the SiV centre. Squares represent the count rates measured on the SiV centre and triangles that beside it. (b) Spectrum of the SiV centre (black line) and of the background (grey line).

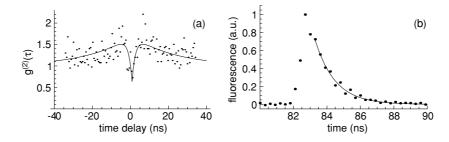


Figure 3. (a) Normalized coincidence rate histogram. The line shows the least square fit using a three-level model. (b) Histogram of the time delay between the laser pulse and the fluorescence detection. The line shows the least square fit of an exponential decay considering the time jitter in the photon detection.

central peak), the counts are due to the dark count rate of the APDs. Additional counts on the right side are mostly due to the Raman scattering of the excitation laser in the bulk diamond. The position of the central peak indicates that this SiV centre lies very close to the surface in agreement with the results of our computer simulation. In figure 2(a), the photon count rate on and aside the SiV colour centre is shown as a function of the pump power. While the background increases linearly with the pump power (grey line), the dependence of the fluorescence on the pump power can be well described by the form $F(P) = F_0 \times P/(P_{\text{sat}} + P)$ (black line), which can be derived for an ideal two-level system. The saturation power P_{sat} was estimated to be 6.87 ± 0.99 mW. Due to the limited power of the excitation laser diode, the saturation behaviour at very strong excitation could not be investigated, which causes the uncertainty in the determination of P_{sat} . Fluorescence spectra were measured at the maximum and at a dark region on the fluorescence image and are shown in figure 2(b). The ZPL of the SiV centre at 738 nm is clearly visible. Due to the low luminescence intensity the vibronic sideband is not resolved.

To evaluate the photon statistics of the fluorescence from the single SiV centres, the autocorrelation function $g^{(2)}(\tau)$ was measured. For this purpose a Hanbury–Brown–Twiss setup consisting of two APDs in each output port of a beam splitter was used. A coincidence unit was used to register histograms of the time delay τ of the detection events of the two APDs [7]. To suppress the background and the optical cross talk between the APDs the interference filter was inserted in front of one APD again.

The normalized photon conincidence rate histogram which is equal to the $g^{(2)}(\tau)$ [20] is presented in figure 3(a). The minimum of about 0.7 at time delay zero proves the nonclassical

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character of the fluorescence. For some τ the value of $g^2(\tau)$ clearly exceeds 1, which indicates an additional shelving state. Similar behaviour has been observed for NV and NE8 centres [7, 11]. To explain the experimental result we employ a three-level model with the two upper levels thermally coupled [7]. Furthermore we assume that background detection events follow a Poissonian distribution. From this model we derive the following autocorrelation function:

$$g^{(2)}(\tau) = 1 + p_f^2(c e^{-\tau/\tau_1} - (1+c) e^{-\tau/\tau_2}), \tag{1}$$

where p_f is the probability that a detection event is due to fluorescence of the single SiV centre. τ_1 is the time constant for photon antibunching and converges to the photoluminescence lifetime for small pump power. τ_2 is introduced due to the shelving effect of the third level. From the least square fit (the line in figure 3(a)) we get the values $p_f = 0.61 \pm 0.18$, $\tau_1 = 1.78$ ns ± 0.64 ns and $\tau_2 = 22.4$ ns ± 5.2 ns. From the saturation measurement we calculated the ratio of fluorescence to total detection events to be about 0.59, which agrees well with the value of p_f . Our results are compatible with perfect photon antibunching. The short value for τ_1 is strongly influenced by the time jitter in photon detection which was separately measured to cause a Gaussian spreading with a FWHM of 770 ps, therefore the relative large error.

To measure the photoluminescence lifetime, the laser diode exciting the colour centres was pulsed with a duration of 200 ps (FWHM) and a repetition rate of 50 kHz. We employed the same coincidence unit used to measure the $g^{(2)}(\tau)$ function to histogram the time delay between the laser pulse and the fluorescence detection. A histogram is presented in figure 3(b), where we used a sample with high density of SiV centres for better statistics. We assume a simple exponential decay including the time jitter in photon detection. From the least square fit shown in figure 3(b) we determined a time constant of 1.20 ± 0.04 ns, in good agreement with the literature value [18].

Considering the short photoluminescence lifetime, much stronger fluorescence should be possible for single SiV centres than observed here. A possible explanation are nonradiative transitions which were reported in investigations of SiV centres in CVD diamonds. There, it was observed that due to these nonradiative transitions the photoluminescence intensity of SiV centres drops at room temperature below 20% of the intensity at 10 K. On the other side, the photoluminescence intensity at 10 K varies over several orders of magnitude for samples grown under different conditions [21]. Probably the nonradiative transitions in the very pure IIa diamonds used here are strongly favoured, resulting in the low photoluminescence intensity of single SiV centres in our samples.

In summary, we have demonstrated the production of single SiV centres in diamond by ion implantation. Ion implantation allows a very precise positioning. This will be crucial for improving the fluorescence collection efficiency of single colour centres using solid immersion lenses, cavities or miniaturizing the setup. In our experiments, a single SiV centre acts as a stable single photon source with narrow emission spectrum at room temperature. All measurements presented here were carried out on the same single SiV centre. Even after three weeks of operation we did not observe any changes in the emission characteristics. Yet, due to the influence of nonradiative transitions, the single photon generation rate still needs some improvements, possibly by modifying the diamond environment. Together with high-Q cavities, high rate emission should be feasible, thereby enhancing the applicability of the SiV centres.

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