Engineering single photon emitters by ion implantation in diamond

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(Received 15 September 2009; accepted 12 October 2009; published online 5 November 2009)

Diamond provides unique technological platform for quantum technologies including quantum computing and communication. Controlled fabrication of optically active defects is a key element for such quantum toolkit. Here we report the production of single color centers emitting in the blue spectral region by high energy implantation of carbon ions. We demonstrate that single implanted defects show sub-poissonian statistics of the emitted photons and can be explored as single photon source in quantum cryptography. Strong zero phonon line at 470.5 nm allows unambiguous identification of this defect as interstitial-related TR12 color center. © 2009 American Institute of Physics. [doi:10.1063/1.3257976]

Single color centers in diamond are continuously attracting attention during the last decade due to their potential application in quantum information science. Several impurity related defects like nitrogen-vacancy complex (NV)^{1,2} and nickel related color center (NE8)^{3–5} were shown to be efficient single photon sources and there are already diamond-based devices on the market.⁶ Furthermore, color centers in diamond are also among the most promising candidates for solid state spin-based quantum computing.^{7–9} NV is a particularly interesting system in that respect owing to its triplet ground state and availability for optically assisted spin read-out technique.^{10,11} The ability to create color centers artificially is of a crucial importance for the above mentioned applications. When combined with diamond nanofabrication techniques,¹² it allows the creation of diamond-based toolbox for quantum communication including single photon sources, optical cavities, and spin-based quantum memories. Nickel related defects were generated in diamond nanocrystals by introducing nickel into the growth medium.^{3,13} However, due to the unpredictable stochastic nature of impurity incorporation in the lattice during growth, such approach suffers from limited accuracy. Recently, ion beam assisted generation of defects was demonstrated for nickel and nitrogen related color centers.^{14,15} This method potentially allows accurate positioning of diamond qubits and is preferable for potential application of single photon sources.

In this letter, we report the selective creation of single color centers in diamond emitting at $\lambda = 470.5$ nm, which is known as TR12 line.^{16–18} It was proposed that this center contains an interstitial carbon atom in a hexagonal site.¹⁹ Although TR12 line has been observed in the luminescent spectrum of diamond decades ago, it was not clear whether luminescence quantum yield is high enough to detect single centers. On the other hand, the simple structure of this defect which does not contain impurity atoms, allows its straightforward fabrication by creation of damage into the diamond. In our study high energy ion implantation was chosen for the creation of vacancies and interstitials. The ion beams with 6 MeV energy used in this study were created by the dynamitron tandem accelerator at the Bochum University and fo-

cused using a 15 T superconducting solenoid lens.²⁰ As a target sample we used a chemical vapor deposition grown diamond with very low nitrogen concentration (<0.1 ppb) provided by Element Six Ltd. After the implantation, the sample was annealed for 2 h at $T = 650$ °C under high vacuum (10^{-7} mbar). It is known that annealing at temperature 400 °C $< T < 650$ °C enhances the creation of the TR12 defect.²¹ In contrast to NV, the TR12 centers are less stable, annealing at temperatures above 800 °C destroys major part of the 470 nm absorption.^{16,22}

Figure 1 shows a confocal microscopy fluorescence image where the implanted pattern can be clearly seen. The ion dose was 2000 ions per spot and each spot in the figure consists of several color centers. Using second-order intensity correlation function measurements we estimated the yield of TR12 formation to be about 0.1% (see below).

In order to prove that the emission from the implanted areas originates from single color centers, we have performed measurements of the photon statistics. Fluorescence light emitted by a single quantum system can be distinguished from the fluorescence of ensemble of atoms by its characteristic subpoissonian photon statistics. When single atom emits a photon, it is projected to the ground state due to energy conservation. When pumped by a coherent (poisso-

FIG. 1. (Color online) Confocal fluorescence image of a pattern of TR12 centers. The excitation wavelength is 473 nm, the detection above 500 nm.

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FIG. 2. Second-order autocorrelation function (antibunching) of a single TR12 center at room temperature.

nian) light source, the fluorescence emission exhibits characteristic antibunching dip in its autocorrelation function. The latter is given by

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

where $I(t)$ and $I(t+\tau)$ are the fluorescence intensities at times t and $t+\tau$, respectively. Experimentally, the photon statistics can be accessed using photon coincidence measurements. In such an experiment the photon stream is divided on a beam splitter and measured by two detectors operating in photon counting mode. The coincidence rate histogram obtained in this way is equivalent to the second-order intensity correlation function for short time scales. Such two detectors set-up avoids dead time problems associated with the response of avalanche photodiodes used in this study.

Experimental data presented in Fig. 2 show subpoissonian photon statistics manifested in a pronounced dip of the fluorescence intensity autocorrelation function at zero delay. Note that $g^{(2)}(\tau)$ shows finite zero delay value. This effect is related to background photons originating from Raman scattering and fluorescing parasitic impurities in the diamond lattice.

Observation of single implanted defects also allows the unambiguous determination of generation efficiency. The contrast of the correlation function gives a direct access to the number of fluorescent defects.²³ By comparing the number of color centers with the number of carbon ions that were implanted, we estimated the yield to be 0.1%. Taking into account that a single carbon ion creates many interstitials and vacancies, this result is surprising. However, there are indication that the structure of the TR12 defect is a complex consisting of interstitials and vacancies.¹⁶ Therefore, the formation of color center is a multistep process where losses of interstitials via recombination with vacancies seriously limit the yield. Further studies for the improvement of the yield of these centers will include optimization of both implantation and annealing procedures.

The above presented results indicate that the emission is originating from a single quantum emitter and can be efficiently used for quantum information processing applications. Apart from the photon statistics, spectral properties of single photon emitters are of primary interest. In order

FIG. 3. Fluorescence spectrum of a single TR12 center at $T=2$ K. The zero phonon line at 470.5 nm belongs to the TR12 defect. The measured line width (half width at half maximum) is 0.14 nm (limited by spectrometer resolution).

to characterize this single photon source, temperature-dependant measurements were performed. Fluorescence spectrum measured at low temperature (2 K) is shown in Fig. 3.

The zero phonon line at 470.5 nm (2.638 eV) and its characteristic squared shape vibronic replica unambiguously show that these defects belong to the TR12 system.¹⁶ Debye-Waller factor characterizing intensity of zero phonon line is moderate (0.1), placed between Ni-related centers (0.5) and nitrogen-vacancy color centers (0.04).

Diamond based single photon source can outperform coherent light sources due to their higher emission rate. The intensity of a single photon source based on single atom emission also has its limitations. The bottleneck defining the maximum number of emitted photons is related to the radiative lifetime of the excited state. In order to characterize this parameter, time resolved fluorescence experiments were performed using pulsed laser as excitation source. Figure 4 shows fluorescence decay of a TR12 defect measured at 1.6 K.

The decay curve is monoexponential with decay constant of 3.6 ns, three times shorter than the decay time observed for nitrogen-vacancy in a bulk diamond.

The results from this study show that interstitial-related TR12 centers are promising candidates for diamond-based

FIG. 4. Fluorescence lifetime measurement of a single TR12 center at $T=2$ K. The solid line is an exponential with decay constant of 3.6 ns corresponding to the lifetime of the excited state.

quantum information processing platform. They show photo-stable emission in the blue spectral region. Little is known about the spin state of this defect. Therefore, both experimental and theoretical work aiming to establish the structure and energy level scheme of this color center is of a crucial importance. Since the structure of this defect consists of carbon atoms and vacancies, the properties related to its nuclear spin can be engineered at atomic level by implantation of different carbon isotopes. Furthermore, recent implantation techniques based on laser cooled ions in Paul trap provide the possibility of deterministic implantation of ions with nanometer accuracy.^{24,25} It is important to note that interstitials related TR12 centers are photochromic.²⁶ Hence our observation of such defects at atomic levels might find an application in atomic memories and super resolution microscopy²⁴

This work was supported by the EU (QAP, EQUIND, NEDQIT, SOLID) DFG, (Grant Nos. SFB/TR21 and FOR730), NIH, Landesstiftung BW, the Volkswagenstiftung, and BMBF (EPHQUAM, KEPHOSI).

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