Stable single-photon source in the near infrared

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New Journal of Physics 6 (2004) 98
Received 1 March 2004
Published 29 July 2004
Online at http://www.njp.org/
doi:10.1088/1367-2630/6/1/098

Abstract. Owing to their unsurpassed photostability, defects in solids may be ideal candidates for single-photon sources. Here we report on generation of single photons by optical excitation of a yet unexplored defect in diamond, the nickel–nitrogen complex (NE8) centre. The most striking feature of the defect is its emission bandwidth of 1.2 nm at room temperature. The emission wavelength of the defect is around 800 nm, which is suitable for telecom fibres. In addition, in this spectral region little background light from the diamond bulk material is detected. Consequently, a high contrast in antibunching measurements is achieved.

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1. Introduction

The technology for creation of single photons is attracting sustained attention because of possible applications in quantum cryptography and computation [1]. Several schemes to produce single photons on demand have been proposed ranging from faint laser pulses to optically excited impurities in solids [2] and quantum dots. Single quantum systems show important advantages in comparison with attenuated classical (Poissonian) sources because of the complete suppression
of multiple-photon emission events. This property of single quantum state emission is related to the influence of the measurement process on the state of a single quantum system. When a photon is emitted by, for example, a single molecule, the system is projected into the ground electronic state. Hence, the simultaneous emission of a second photon is impossible. In the recent past, several such systems have been proposed as single-photon emitters. First attempts have been carried out on single molecules at low temperature [3] as well as under ambient condition [4, 5]. In the latter case, a major drawback is the limited photostability of organic dye molecules at room temperature, whereas in the first case, liquid-helium temperatures are prohibitive for most practical applications. Nevertheless, molecules are still interesting candidates for single-photon sources mainly because they allow for flexibility in the choice of emission wavelength. Alternatively, quantum dots have been proposed as emitters. For this case, electrical and optical pumping of single-photon emission has been shown [6]–[8]. A certain disadvantage is the limited emission wavelength and the requirement of low-temperature operation. The only photostable single-photon source at room temperature reported so far is the nitrogen-vacancy defect centre in diamond [9]–[11]. A serious disadvantage of the N–V defect is its spectrally broad emission band and also the emission wavelength, which is around 640 nm.

It is known that certain types of defects in diamond, in particular when they are not associated with a vacancy, show a much narrower emission range [12]. In the following, we will describe such a defect. Recently, several new defects emitting in the infrared spectral region which are related to Ni ions have been described in the literature [13]–[15]. It should be noted that the absorption of the Ni-related centres in natural diamond is more than one order of magnitude weaker than in synthetic stones. This is because untreated stones contain defects related to impurity in this ion in very low concentration.

2. Experimental

For experiments, natural IIa type diamonds (Drukker), without special treatment, were used. Single defects in diamond had been selected using a conventional confocal microscope. Excitation was carried out either by a pulsed laser (Coherent Mira; repetition frequency 76 MHz and excitation wavelength 695 nm) or continuously with an excitation wavelength of 710 nm. The fluorescence light from a single-defect centre was collected and directed to the detection channel, which includes a spectrometer and a Hanbury-Brown and Twiss interferometer. Photon arrival events were analysed using a single-photon counting card (SPCM 730, Becker&Hickl). An electronic delay was inserted into one of the detection channels in order to check the symmetry of the autocorrelation function. The fluorescence emission spectrum was detected with a 0.3 m imaging spectrometer (Acton research) equipped with a back-illuminated CCD camera (Roper Scientific).

3. Results and discussion

Less than 30% of the untreated IIa diamonds showed a concentration of NE8 defects suitable for single-centre detection using confocal microscopy (∼1 defect/100 μm³ structure; see figure 1). The NE8 defects were homogeneously distributed over the whole diamond sample. It should be noted that some of the IIa diamonds did not show any NE8 defects. The fluorescence emission spectrum obtained from a single-defect centre is shown in figure 2. The spectrum shows one
pronounced zero-phonon line at 802 nm, together with a shallow phonon side wing extending up to 850 nm. The relative integral intensity of the zero phonon line to the entire spectrum (Debye–Waller factor) is 0.7, higher than the corresponding Debye–Waller factor for e.g. the N–V centre in diamond which is 0.04. Time-resolved fluorescence decay measurements (data not shown) show a monoexponential decay curve with a decay constant of 11.5 ns.

Fluorescence intensity autocorrelation measurements have been performed to demonstrate the sub-Poissonian statistics of the light emitted by the centre. Using a start-stop scheme, the coincidence rate between the two detectors of a Hanbury–Brown and Twiss interferometer has been measured. The data obtained are equivalent to the second-order intensity autocorrelation function for short time scales. The normalized autocorrelation function \( g^{(2)}(\tau) = \langle I(t)I(t+\tau) \rangle / \langle I(t) \rangle^2 \) was obtained from a photon coincidence rate histogram [16].

Data are presented in figure 3. The autocorrelation function shows a pronounced minimum at zero delay time \( \tau \). The remaining difference from zero at \( \tau = 0 \) results from background.
light, mostly related to Raman scattering from the diamond lattice. In addition to antibunching, the autocorrelation function also shows photon bunching \( g^{(2)}(\tau) > 1 \). Such a behaviour was previously reported for single organic molecules and the N–V centre and is related to an additional metastable state in the photoexcitation cycle. Photon bunching is a measure of the lifetime of the metastable level and the intersystem crossing (ISC) rate. This can be understood as follows. Time traces of a three-level quantum system, pumped by an excitation source between levels 0 and 1, consist of bright (fluorescence) and dark (no fluorescence) intervals. The bright intervals correspond to the evolution of the system between the states 0 and 1 (see figure 1). If the system undergoes a transition to the metastable state, it will not fluoresce and a dark interval will occur with a length equivalent to the lifetime of the metastable state.

In order to determine the ISC rates, a theoretical model had been used to fit the data [17]. The evolution of the system can be described by optical Bloch equations for a three-level system (figure 1). In general, such a system can only be solved numerically. However, due to the high dephasing of the optical transition at room temperature (\( \approx 400 \) GHz), the optical Bloch equations can be reduced to rate equations and solved analytically:

\[
\dot{\rho}_1 = -(\Gamma + k)\rho_1 + k\rho_0, \\
\dot{\rho}_0 = \left(\frac{1}{T_1} + k\right)\rho_1 - k\rho_0 + \gamma_{20}\rho_2, \\
\dot{\rho}_2 = \Gamma_{12}\rho_1 - \gamma_{20}\rho_2, \\
\]

where \( \Gamma = (1/T_1) + \Gamma_{12} \), with \( \Gamma_{12} \) being the ISC rate, \( k \) the absorption rate and \( \gamma_{20} \) the decay rate of the metastable state. The normalized autocorrelation function is \( g^{(2)}(t) = p(t)/p(\infty) \), where

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure3}
\caption{The fluorescence intensity autocorrelation function measured with cw excitation; the experimental data are shown as circles, while the red line represents the theoretical curve, calculated according to the procedure described in the text. The autocorrelation function shows a good contrast and presents a pronounced dip close to \( t = 0 \).}
\end{figure}
Table 1. The fit parameter values from the autocorrelation function.

| Symbols | Meaning                              | Value (MHz) |
|---------|--------------------------------------|-------------|
| $k$     | Absorption rate                      | 440         |
| $1/T_1$ | Relaxation rate of the excited state | 87          |
| $\Gamma_{12}$ | ISC rate                  | 17          |
| $\gamma_{20}$ | Decay rate of the metastable state    | 6.1         |

Figure 4. The fluorescence intensity saturation curve; symbols represent the experimental data and the red line a fit to the data according to the formula in the text.

$p(t)$ is the analytical solution for the two-photon correlator:

$$p(t) = \frac{k}{T_1} \left[ \frac{\gamma_{20}}{\gamma_0 - R^2} + \left( 1 - \frac{\gamma_{20}}{\gamma_0 - R} \right) e^{-\gamma_0 - R t} \right] - \left( 1 - \frac{\gamma_{20}}{\gamma_0 + R} \right) \frac{e^{-\gamma_0 + R t}}{2 R},$$

with

$$\gamma_0 = \frac{\Gamma + 2k + \gamma_{20}}{2} \quad \text{and} \quad R = \sqrt{\left( \frac{\Gamma + 2k - \gamma_{20}}{2} \right)^2 - \Gamma_{12} k}.$$ 

The autocorrelation curves were fitted with (2), using $\Gamma_{12}$ and $\gamma_{20}$ as fit parameters (figure 3). The fit values are shown in table 1.

In order to determine the maximum emission rate and to derive the fluorescence quantum yield, the fluorescence saturation curve was recorded (figure 4). In the low laser power regime, the fluorescence intensity linearly increases with laser power. For high laser powers, the population probability of the metastable level increases, and the fluorescence intensity saturates. The saturation data were fitted using $R = R_\infty (I/I_5)/(1 + I/I_5)$, where $I_5$ is the laser power corresponding to saturation and $R_\infty$ is the fluorescence intensity corresponding to infinite laser power. The fully saturated signal $R_\infty$ in our experiments was 75 000 counts s$^{-1}$. However, in order...
to obtain the emission rate of the centre, the set-up detection efficiency must be taken into account. The detection yield is limited by the transmission through the objective ($\eta_1 \approx 0.8$), the surfaces of the optical set-up (lenses, beam splitter and filters, $\eta_2 \approx 0.2$), the quantum efficiency of the detector ($\eta_3 \approx 0.7$), first-order diffraction maximum intensity of the microscope lens ($\eta_4 \approx 0.8$), and light collection efficiency of our microscope objective for transition of the centre in a high refractive medium ($\eta_5 \approx 0.07$). The resulting efficiency is therefore $\eta_{\text{res}} = \eta_1 \eta_2 \eta_3 \eta_4 \eta_5 \approx 0.5\%$. Taking this into account, the photon emission rate is $15 \times 10^6$ counts s$^{-1}$. This photon emission rate is smaller than that for an ideal photon emitter, i.e. a two-level system. Using the ISC rates specified above, the maximum emission rate for a three-level system can be calculated\textsuperscript{[18]} as

$$R_\infty = \frac{(1/T_1 + \Gamma_{12}) \Phi_F}{2 + (\Gamma_{12}/\gamma_{20})},$$

where $\Phi_F$ is the fluorescence quantum yield. With an estimate detection efficiency of around 0.5%, a fluorescence quantum yield of $\Phi_F = 0.7$ results. For $\Gamma_{12} = 0$, a two-level maximum rate is obtained. With the rates obtained (see table 1), the ratio between the maximum emission rates for three- and two-level systems is 0.5, i.e. the measured value for the fluorescence intensity is roughly 50% of the maximum possible one (for a two-level system). This high value is due to the fact that the ISC rate is low compared with the radiative emission rate. In addition, the decay rate of the metastable state is high, hence the photon counting rate is comparable with that of the N–V centre.

4. Conclusions

We report on a new candidate for a single-photon source based on the single NE8 defect in diamond. This defect shows an intense and spectrally narrow emission, suitable for generation of transform-limited single-photon wavepackets, which is important for optical quantum computing schemes\textsuperscript{[19]}. Furthermore, the emission is in the near infrared, making it important for fibre-based quantum cryptography schemes.

Acknowledgments

This work is supported by the DFG Schwerpunktprogramm ‘Quanten-Informationsverarbeitung’.

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