A highly efficient two level diamond based single photon source

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An unexplored diamond defect centre which is found to emit stable single photons at a measured rate of 1.6 MHz at room temperature is reported. The novel centre, identified in chemical vapour deposition grown diamond crystals, exhibits a sharp zero phonon line at 734 nm with a full width at half maximum of \( \sim 4 \) nm. The photon statistics confirm the center is a single emitter and provides direct evidence of the first true two-level single quantum system in diamond.

Nanotechnology and photonics industries around the world are currently undergoing a revolution as research into individual quantum systems opens up new and exciting opportunities in quantum information processing (QIP) and quantum key distribution (QKD). At the forefront of this revolution is the development of light sources which emit single particles of light on-demand, known as single photon sources (SPSs). Single photon generation has been demonstrated with several different technologies including quantum dots (QDs), single molecules and more recently carbon nano-tubes, but perhaps the most promising and applicable source of single photons for practical applications arise from optical defects in diamond which can be easily accessed and operate at room temperature.

To date, of the over 500 known defect centres in diamond only three have been identified as single photon (SP) emitters. However, what sets these colour centres apart from the other technologies is their uncompromised photo-stability at room temperature. Until now, the SP emission rates achieved in diamond based SPSs have been limited to \( \sim 300 \) kHz due to the presence of meta-stable/shelving states which act to reduce the efficiency of the single quantum system. In this paper we report the first diamond based defect centre which behaves as a true two-level system. This centre also exhibits the highest measured SP emission rate at room temperature in the MHz regime.

Many schemes have been proposed to improve the emission rate from diamond based SPSs with perhaps the most promising involving the incorporation of single emitting diamond nano-crystals into cavity structures. Unfortunately, the lack of mature cavity technology has limited the incorporation of diamond based SPSs into such designs. As a result, researchers are in parallel exploring new defect centres which may result in more efficient SP emission. For diamond based SPSs to emit more efficiently new defect centres need to be explored and/or created which comprise a simple two level energy structure similar to that observed in semiconductor QDs. To this end, this work explores novel defect centres which can be created synthetically using the well known microwave plasma enhanced chemical vapour deposition (MPECVD) technique.

The fused silica substrate characterised in this work was \( 1 \times 1 \) \( \mu m \) in size and ultrasonically seeded with commercial grade diamond powder 0-2 \( \mu m \) in size prior to MPECVD growth. The 30 minute diamond growth was conducted under the conditions described in [15], with background levels of nitrogen, nickel and silicon present in the system. Photo-luminescent studies were conducted on the fused silica substrate with the aim of identifying novel luminescent defect centres. Many emitting centres were identified by the in house fibre based scanning confocal microscope, operating with 532 nm excitation and a spatial resolution of \( \sim 400 \) nm. Figure 1 (a) illustrates the large number of emitting centres within a \( 20 \times 20 \) \( \mu m \) area of the substrate. The photo-luminescence (PL) spectra from each emitting crystal was measured in the first instance to determine the type of emitting centre and secondly to identify any uncharacteristic spectral lines. The room temperature PL spectrum from the emitting centre identified in the centre of Figure 1 (b) is shown in Figure 1 (c). The spectra reveals a sharp zero phonon line (ZPL) centered at 734 nm with a full width at half maximum (FWHM) of 4.1 nm. The Huang-Rhys factor which is a measure of the relative intensity of the ZPL compared to the entire spectrum was found to be 0.81 which is higher than that reported for the nickel related (NE8) centre and significantly higher than the well known nitrogen vacancy (N-V) centre \( 0.04 \). The photon statistics of the centre were investigated using a fibre based version of the Hanbury Brown and Twiss interferometer. The second-order intensity autocorrelation function:

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

obtained from the photon coincidence rate histogram is shown in Figure 2 for three different incident pump powers. The autocorrelation function minimum at the zero delay time, \( g^{(2)}(0) \), was measured to be 0.16, with the deviation from zero being attributed to the background surrounding and/or within the emitting crystal. The important conclusion to be drawn from the
autocorrelation measurements, as a function of pump power, is the absence of 'photon bunching' [17, 18] in the coincidence histogram. Photon bunching is observed in all of the single diamond related defects reported to date [8, 9, 10] and is attributed to the presence of a meta-stable quenching/shelving state in the electronic structure of the atomic system. The absence of photon bunching in the coincidence histogram for excitation powers well above saturation is direct evidence of a two level energy scheme. The power dependence of the second order correlation function based on a two level energy scheme can be described by [9]:

\[
g^{(2)}(\tau) \simeq 1 - \exp(-k_T \tau) 
\]

where \(k_T = k_{21} + k_{12}\), with \(k_{12}\) and \(k_{21}\) being the pumping rate and photon emission rate of the excited state, respectively.

A least squares fit of Eq. 1 was applied to the measured anti-bunching data as a function of pump power, as seen in Figure 2. The fitting parameter \(k_T\) can then be used to obtain the fluorescence decay rate of the excited state as \(k_T(0) = k_{21}\), in the limit of zero pump excitation \((P \rightarrow 0)\). Figure 3 shows the fitting parameter \(k_T\) as a function of excitation power. From the applied linear fit to the data the excited state lifetime \(\tau_2 = 1/k_{21} = 13.6\) ns. This lifetime is comparable to the (N-V) centre in bulk diamond (11 ns) [19] and considerably longer than those observed from the (NE8) (2 ns) [17] and silicon-vacancy (Si-V) (1.2 ns) [10] centres; however due to the two level nature of the atomic centre it is able to emit more efficiently compared to its three level system counterparts. This is evidenced by the extraordinary single photon emission rate (see Figure 4) measured as a function of pump power, under 532 nm
excitation. The saturation behaviour of the emission

is consistent with that expected for a single atomic
system with a finite lifetime. The theoretical fit to the
measured emission data was derived from the steady
state solution of the excited state population of a two
level system and is given by: $I(P) = I_{sat} P / (P_{sat} + P)$,
where $I_{sat}$ represents the photon saturation count rate,
P is the incident pump power and $P_{sat}$ is the saturation
pump power. From the fit to the experimental data
$P_{sat} = 224$ µW and $I_{sat} = 1.8$ million counts/s. The
saturation power is comparable to that found for N-V
centres and considerably less than that observed for
the NE8 and Si-V centres. Moreover, the saturation
count rate of the 734 nm centre is the highest reported
for a single diamond defect and the first observation
of stable MHz emission from a single photon source
operating at room temperature. The two level nature of
the centre also allows an accurate estimate of the total
collection efficiency of the system to be made as there
is no quenching state reducing the number of emitting
photons. Based on the fluorescence lifetime, the centre
emits at 73.5 MHz of which 1.6 MHz was detected;
this equates to a total system collection efficiency of
2.17%. With further optimization of the substrate and
the potential to manipulate diamond crystals onto
plasmonic structures \cite{19}, there is the possibility to
achieve SP emission into the tens of MHz range at room
temperature.

The atomic structure of the single emitting defect
centre is at this stage unclear, there are reports of a 733
nm absorption peak from Si related centres in single
crystal diamond, however no emission peaks from the
733 nm line have been observed in the PL under 514 nm
excitation \cite{21}. One can speculate though that the origin
of the centre lies within a mixture of the constituents
present in the diamond reactor during growth which
includes nitrogen, silicon, nickel and hydrogen. PL stud-
ies of the diamond seed material before growth under
532 nm excitation revealed N-V centres were present
in a number of seed crystals, however, this fluorescence
emitter was only observed after being exposed to the
MPECVD process.

A new and highly efficient diamond-based single
photon source operating in the infra red at 734 nm has
been fabricated and characterised. The diamond defect
exhibits an intense and spectrally narrow emission line
and is the first SPS to exhibit MHz operation at room
temperature. The centre is found to significantly extend
the performance range of diamond based SPSs due to its
two level nature. With the opportunity to manipulate
such sources onto structures, which can further enhance
the emission properties, the future for practical room
temperature quantum devices looks bright.

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\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Photon emission rate for the 734 nm single emitting
centre as a function of pump power when excited at 532 nm.
The emission rate is given by the sum of the counts on the two
APDs in the Hanbury Brown and Twiss arrangement. The fit
to the measured data is discussed in the text. The measured
emission rates for a single N-V and NE8 centre are included
in the figure as a comparison and were all measured using the
same experimental setup, with the exception of the NE8 data
which was measured under 685 nm excitation.}
\end{figure}

[1] M. Nielsen et al., Quantum computation and quantum
information. (Cambridge University Press, Cambridge
1964).
[2] J. Wrachtrup et al., J. Phys.: Condens. Matter 18, 807
(2006).
[3] A. Greentree et al., Mater. Tod. 11, 22 (2008).
[4] N. Gisin et al., Rev. Mod. Phys. 74, 145 (2002).
[5] C. Santori et al., Phys. Rev. Lett. 86, 1502 (2001).
[6] C. Brunel et al., Phys. Rev. Lett. 83, 2722 (1999).
[7] A. Hogele et al., Phys. Rev. Lett. 100, 217401 (2008).
[8] T. Gaebel et al., New J. Phys. 6, 98 (2004).
[9] C. Kurtsiefer et al., Phys. Rev. Lett. 85, 290 (2000).
[10] C. Wang et al., J. Phys. B: Atomic molecular and optical
physics 39, 37 (2006).
[11] J. Rabeau et al., Appl. Phys. Lett. 86, 131926 (2005).
[12] I. Aharonovich et al., Appl. Phys. Lett. 93, 243112
(2008).
[13] C. H. Su et al., Opt. Exp. 16, 6240 (2008).
[14] S. Strauf et al., Nature Photon. 1, 704 (2007).
[15] J. Rabeau et al., Appl. Phys. Lett. 86, 134104 (2005).
[16] R. Twiss et al., Nature 180, 324 (1957).
[17] E. Wu et al., Opt. Exp. 14, 1296 (2006).
[18] F. Jelezko et al., J. Phys. Stat. Sol. A 203, 3207 (2006).
[19] A. Beveratos et al., Phys. Rev. A 64, 061802 (2001).
[20] A. Collins et al., Nature 450, 402 (2007).
[21] I. Kiiflawi et al., Diamond Relat. Mater. 6, 146 (1997).