



Instabilities in nanodiamond nitrogen-vacancy centre single photon sources under prolonged pulsed excitation

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Abstract: Colour centres in nanodiamonds provide robust sources of fluorescence and can be used as triggered sources of single photons at room temperature. However, practical devices require stability over thousands of hours of operation, and the use of strong pulsed optical excitation, placing significant burden on the robustness of the emitters that requires bespoke testing. In this work we report the response of single NV centres in nanodiamonds of 50 nm and 100 nm diameter to accelerated lifetime testing, exciting the defects close to saturation around 10^{13} times to simulate the minimum operational lifetime of a practical device. For nanodiamonds 50 nm in diameter, observed changes in the fluorescence intensity and lifetime suggest a progressive size reduction as a result of the pulsed laser excitation, combined with the introduction of non-radiative centres on or near the nanodiamond surface which affect the quantum efficiency of the NV centre and ultimately lead to photobleaching of the emission. We find examples of NV centres in 100 nm nanodiamonds for which triggered single photon emission remains stable for over these accelerated lifetime tests, demonstrating their suitability for use in practical devices.

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1. Introduction

Colour centres in diamond show promise for use in a range of quantum photonic devices including single photon sources. Several different colour centres [1–3] have been investigated which show potential as photostable, room-temperature, single photon emitters [4]. The nitrogen vacancy (NV) centre has received particular interest [5,6] owing to its near-unity quantum efficiency in bulk diamond, high photostability and easy optical pumping. However the NV centre suffers from low spectral density of emission, with a broad phonon sideband with full-width at half-maximum 660–720 nm, which is undesirable for many applications. Coupling the emission to an optical microcavity mode [7] can significantly increase the spectral density of the source, but brings with it substantial challenges in the reproducible engineering of robust devices.

A significant aspect of this challenge is the need for micro or nanoscale diamond structures in order to place the NV centre in a microcavity. An NV centre within a nanodiamond experiences a lower photonic density of states than in the bulk [8], leading to a longer radiative lifetime [9–12] and reduced quantum efficiency [12–14]. In the smallest nanodiamonds reduced photostability is observed as a result of additional non-radiative relaxation pathways [15].

Despite extensive work on the characterisation of emission from NV centres in nanodiamond and their suitability for practical single photon sources, little has been reported on their robustness to operating conditions of devices. Colour centre single photon sources are generally excited optically with intense laser pulses, such that degradation can potentially occur due to photophysical and photochemical changes to the material and its environment and, in extreme situations, to

the defect structure itself. Such photodegradation effects of spectral diffusion, blinking and photobleaching are well-known in optically excited fluorophores including dye molecules [16] and in nanomaterials such as colloidal quantum dots [17]. Despite NV centres in diamond being the most robust optically excited single photon emitters known, the substantial performance requirements of practical devices make this topic worthy of attention. High efficiency non-resonant excitation of the NV centre requires pulsed excitation fluence of order 10 mJ cm^{-2} , while repetition rates of order 5 MHz combined with operational lifetimes of 1000 hours suggest in excess of 10^{13} excitation cycles, such that the nanodiamond host is exposed to over 100 GJ cm^{-2} of excitation energy over its operational lifetime.

Here we present a study of the effects of prolonged laser excitation on single photon emission from NV centres in nanodiamond. Following extended periods of 532 nm pulsed excitation, NV centres in 50 nm nanodiamonds displayed decreased quantum efficiency, decreased emission stability and increased measured decay lifetime. These changes, we suggest, originate from a gradual reduction in nanodiamond size over the excitation periods as a result of laser-induced nano-etching of the diamond surface. By contrast, emission from NV centres in 100 nm diameter nanodiamonds was observed to be stable over the extended excitation period. We discuss these results in the context of nanodiamond based photonic devices.

2. Experimental methods

High-pressure high-temperature (HPHT) nanodiamond samples with mean diameters of $50 \pm 15 \text{ nm}$ and $100 \pm 25 \text{ nm}$ [18] were purchased from FND Biotech. Approximately 10% of nanodiamonds are single NV centre crystals, demonstrating the strong photon anti-bunching shown in Fig. 2(A). Nanodiamond solution of concentration 0.1 mg/ml was spin-coated, $100 \mu\text{L}$ at a time at 1200 rpm, onto a planar substrate coated with a dielectric mirror until the desired coverage was achieved (4-8 coats). The mirror, produced by LayerTec GMBH, consisted of 7 pairs of SiO_2 and Ta_2O_5 , giving a peak reflectivity of 97.5% at 690 nm. The mirror was patterned with a focused ion beam to provide registration marks allowing relocation of emitters for different parts of the experiments. The use of mirrors is in preparation for cavity coupling, as reported elsewhere [7].

The samples were characterised using a home-built confocal microscope with an objective lens numerical aperture of 0.85 (Olympus LCPLFLN100xLCD). Two excitation sources at 532 nm were used, a continuous wave laser (Oxxius LCX-532L 532 nm 50 mW), and a pulsed laser (PicoQuant LDH-PFA-530B 532 nm). The pulsed laser has a 100 ps pulse duration, providing a measured pulse energy of 32.3 pJ with a variable repetition rate up to 80 MHz. Both lasers were focused onto the sample as diffraction-limited spots, giving a fluence of 11 mJ cm^{-2} . As shown by Fig. 2(B), this pulse energy is sufficient for NV centre emission saturation. An external trigger was used to control laser driver (PicoQuant PDL 800-B), giving software control the of repetition rate and exposure time. Nanodiamonds with single NV centres were selected for investigation. Time-resolved photoluminescence (TRPL) and Hanbury Brown and Twiss (HBT) measurements were recorded with single photon counting detectors (Excelitas) and time-correlated single photon counting electronics (Swabian Instruments TimeTagger 20). Spectra were taken with a spectrometer (Andor Kymera 193I-A) equipped with a Peltier-cooled CCD (Newton DU940P-BV).

For accelerated lifetime testing (ALT) selected NV centres were subject to 80 MHz pulsed laser excitation over 62.5 hours, giving an equal pulse number to operation at 5 MHz repetition rate over 1000 hours - a reasonable minimum viable device lifetime. NV2 was tested for an extended time (140 hours) to explore the full extent of degradation behaviour. Such a mapping assumes that degradation takes place on a 'per pulse' basis independent of repetition rate, which is a reasonable assumption given the fast relaxation rates on the diamond surface under ambient conditions. We note also that any memory effects between pulses would be likely to increase the rate of degradation by introducing a cumulative element to the energy delivered, such that

stability under ALT would provide a conservative measure of the operational lifetime of the device.

During the ALT, laser alignment was maintained using a 3-axis Gaussian peak-finding realignment routine running at four minute intervals. Figure 1 provides an outline of measurement sequencing. Spectrograph and HBT measurements were taken at 4 minute intervals, under 80 MHz excitation, for 180 seconds of exposure. TRPL measurements, which measure the decay dynamics of the NV centre, were performed at 20 minute intervals (1 in 5 measurement cycles) with an exposure time of 30 seconds. The laser excitation rate was reduced to 5 MHz during the measurement period in order to give the NV centre sufficient time to decay before subsequent excitation (97.8% recovery for $\tau = 50$ ns). The mean count rate was also recorded during the 5 MHz excitation period to monitor the system quantum efficiency from which changes to the NV emission efficiency could be inferred. TRPL data were fitted with a bi-exponential function with a constant background component using a non-linear least-squares method.

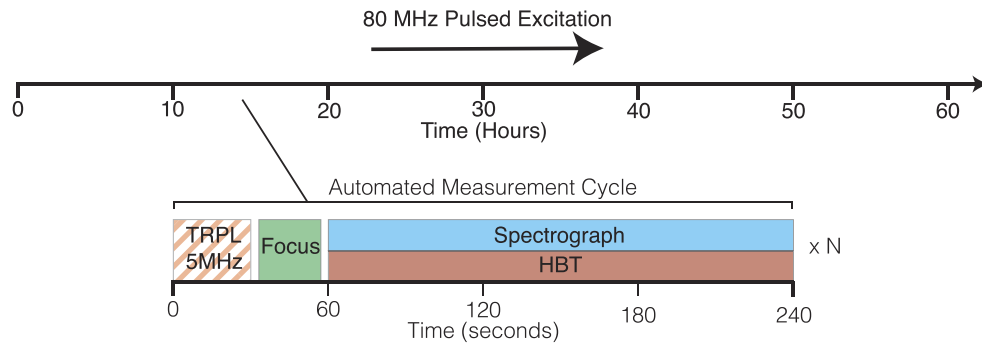


Fig. 1. Schematic outlining accelerated lifetime testing measurement cycle. 30 second 5 MHz time-resolved photo-luminescence measurement performed at 20 minute intervals, Gaussian peak-find focus, Hanbury Brown and Twiss measurements, and Spectrographs performed at 4 minute intervals.

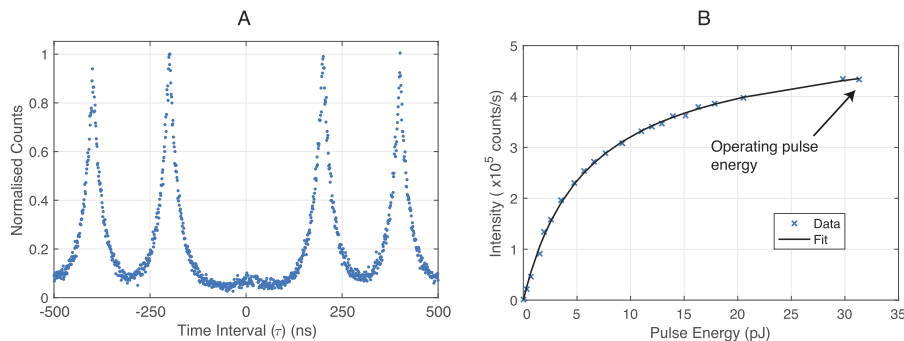


Fig. 2. Single photon emission under pulsed excitation. A: 5 MHz pulsed photon correlation measurement of a single NV centre in nanodiamond showing strong photon anti-bunching. B: Measured fluorescence intensity as a function of laser power at 80 MHz, consisting of linear background and asymptotic NV response. The excitation power used is sufficient to achieve NV centre emission saturation.

3. Results

Prior to extended exposure there was significant range of NV centre brightness (count rate) and emission stability (second-by-second variation in brightness) under both continuous and pulsed excitation and measured decay lifetimes - see Fig. 3. The range of all three parameters is observed to be broader in the 50 nm sample than the 100 nm sample. The most stable nanodiamonds in both samples show less than 1% variation in emission brightness under continuous wave excitation, whereas the least stable have sporadic sharp brightness drops down to a fraction of the initial brightness value.

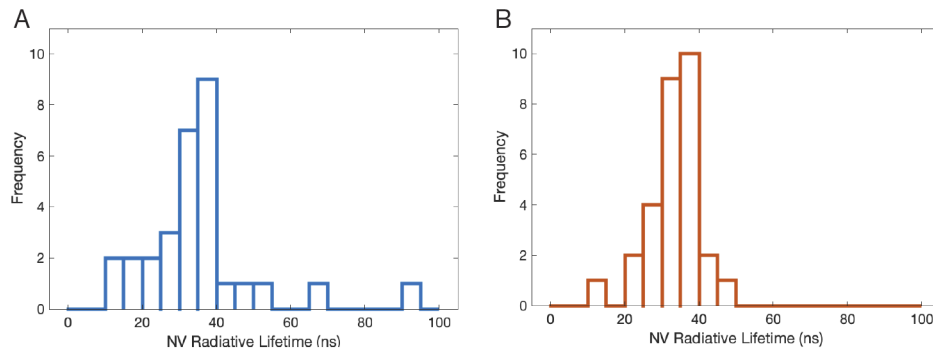


Fig. 3. Lifetime distributions of nanodiamond samples before extended exposure. A: 50 nm sample, $n=30$, mean=34.8ns, range=77.1 ns. B: 100 nm sample, $n=29$, mean=32.6 ns, range=33.2 ns.

3.1. Long-term stability of NV centres in 50 nm nanodiamonds

Over periods of prolonged pulsed exposure, changes in the intensity and lifetime of the fluorescence were observed but there were no noticeable changes in the emission spectra that would indicate instability in the NV charge state via the formation of sp^2 bonded carbon at the nanodiamond surface [19,20]. Likewise we did not observe a significant increase in $g^2(0)$ which would indicate the generation of additional NV centres within the nanodiamond [21]. Accelerated lifetime tests were carried out on seven 50 nm diameter nanodiamonds, revealing a range of behaviour with some showing greater long-term stability than others. Data from the least stable (and longest lifetime) NV measured (NV1) is shown in Fig. 4. Prior to prolonged pulsed excitation this NV centre had a very long measured lifetime of 90.1 ns suggesting that the NV centre is situated in a small nanodiamond and that the relaxation is primarily radiative. The lifetime and count rate during 5 MHz excitation both show a progressive reduction over a period of two hours. The changes in measured lifetime correlate well with the changes in measured brightness, consistent with an increasing non-radiative relaxation rate. Emission ceases after two hours and does not recover. As well as the slow changes shown in Fig. 4, emission from this NV centre showed sudden jumps in intensity second-by-second which became more severe over the course of the excitation period.

The most stable NV centre of those investigated from the 50 nm sample (NV2) revealed quite different behaviour, as shown in Fig. 5. This NV was exposed well beyond the 62.5 hour lifetime equivalent to test the full extent of its stability. The initial lifetime of NV2 was measured to be 48 ns, which is comparably high, with a similar initial count rate to NV1 suggesting that the NV2 host nanodiamond is larger. During the first 36 hours of excitation under the same conditions, only a slight reduction in lifetime to 46 ns is observed, with the brightness staying roughly constant through this period. Between 36 and 56 hours the lifetime drops markedly to 35 ns with a concurrent drop in brightness. Then between 56 and 140 hours the lifetime shows a gradual

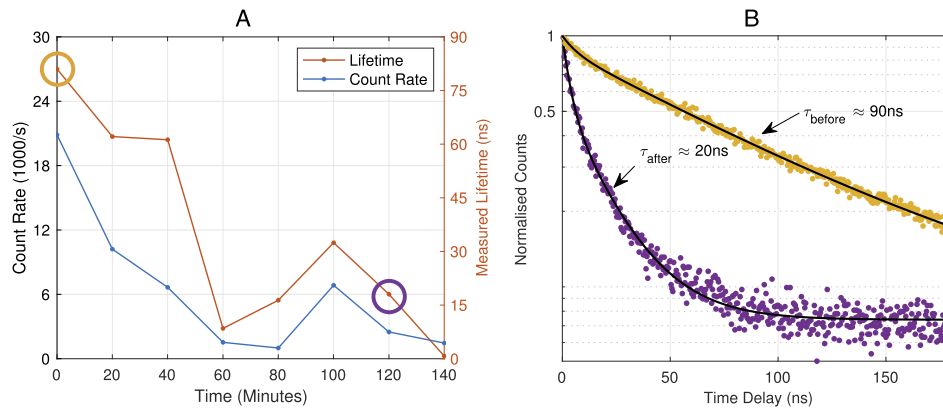


Fig. 4. Fluorescence decay dynamics of a 50 nm nanodiamond NV1 A: Variation in count rate and lifetime over 140 minutes of 80 MHz pulsed exposure revealing reduction in both lifetime and count rate. Measurement cycle outlined in Fig. 1. B: TRPL curves before (yellow) and after (purple) 120 minutes of pulsed excitation. $\tau_{\text{before}} = 90.1$ ns, $\tau_{\text{after}} = 20.0$ ns.

(although noisy) increase to about 60 ns whilst the brightness stays roughly constant. Short term switching in brightness similar to that of NV1 was observed from about 50 hours onward.

Over the full 140 hour excitation period NV2 showed a 29% increase in the measured decay lifetime from 47.7 ns to 61.5 ns and a progressive increase in variation between successive measured lifetimes. TRPL measurements sample just 30 seconds of NV centre emission, which varies on a comparable timescale, therefore substantial variation is observed between successive lifetime measurements.

Before-after analysis of a similarly stable NV showed similar behaviour to NV2. Following 62.5 hrs of exposure at 80 MHz, a 19% increase in measured lifetime of 38.8 ns to 46.2 ns, a 36.2% decrease in continuous-wave saturation count rate from 273 kcounts/s to 174 kcounts/s and a 57.8% decrease in count rate under 5 MHz pulsed excitation from 64.4 kcounts/s to 27.2 kcounts/s were observed. The other 50 nm nanodiamonds studied showed behaviour between these extremes, with progressively less stability resulting in eventual quenching of emission.

The measured lifetime is the product of the pure radiative lifetime with the quantum efficiency of the emission process ($\tau = \eta \times \tau_{\text{rad}}$) [12]. Dividing the measured lifetime by the count rate during the TRPL measurement (5 MHz excitation) therefore gives a value proportional to the radiative lifetime, as shown in Fig. 5(B). After 40 hours, a steady increase over the exposure period can be observed. A similar trend was observed in all 50 nm samples investigated. The substantial scatter on the plot can be attributed to variation in the quantum efficiency during individual TRPL measurements (main cause of outliers), inaccuracy in lifetime fitting ($\approx \pm 5\%$), variation in the laser focus ($\approx 20\%$) and potential changes in collection efficiency due to rotation of the nanodiamond.

3.2. Short-term fluorescence instability of NV centres in 50 nm nanodiamonds

To investigate further the short term emission instability dynamics shown by NV1 throughout and NV2 after 50 hours of excitation, NV2 was subject to the measurement cycle outlined in Fig. 6(C). 184 TRPL measurements, each with an exposure time of 30 seconds under 5 MHz pulsed excitation, were taken successively over a period of three hours whilst the fluorescence count rate was simultaneously recorded. TRPL curves were fitted using the same method outlined previously, with automated alignment in the intervals between measurements. The radiative

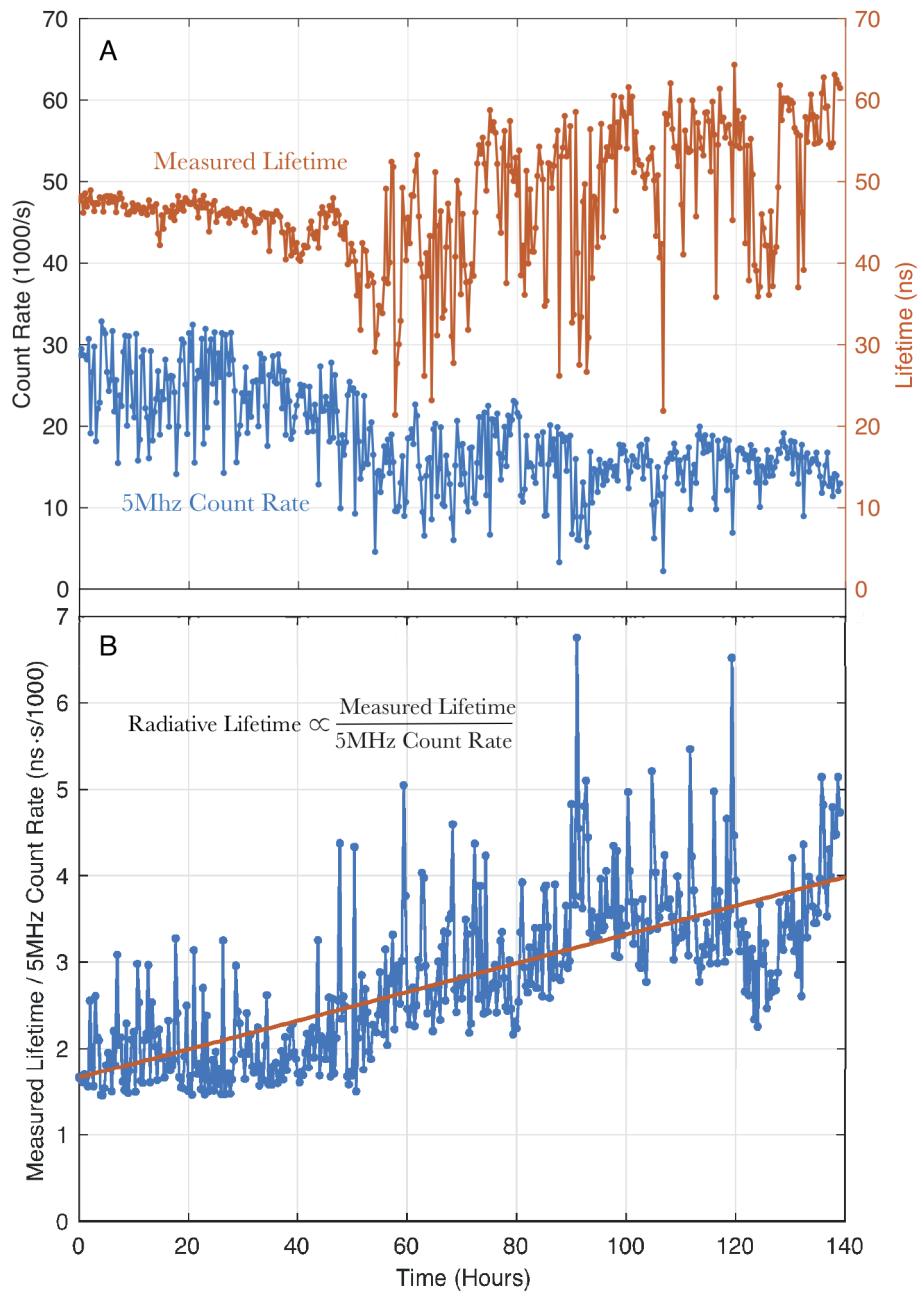


Fig. 5. Fluorescence decay dynamics of a 50 nm nanodiamond NV2. A: Variation in count rate and lifetime over 140 hours of 80 MHz pulsed exposure. Demonstrates comparably greater stability compared to NV1. Measured lifetime increases overall from 47.7 ns to 61.5 ns, 5 MHz count rate decreases by over 50%. Increase in emission instability over time (seen as scatter). B: Division of measured lifetime and count rate, approximately proportional to radiative lifetime, which progressively increases over experiment duration.

lifetime can be assumed to be constant over the three hour period given the slow rate of change of this parameter presented above.

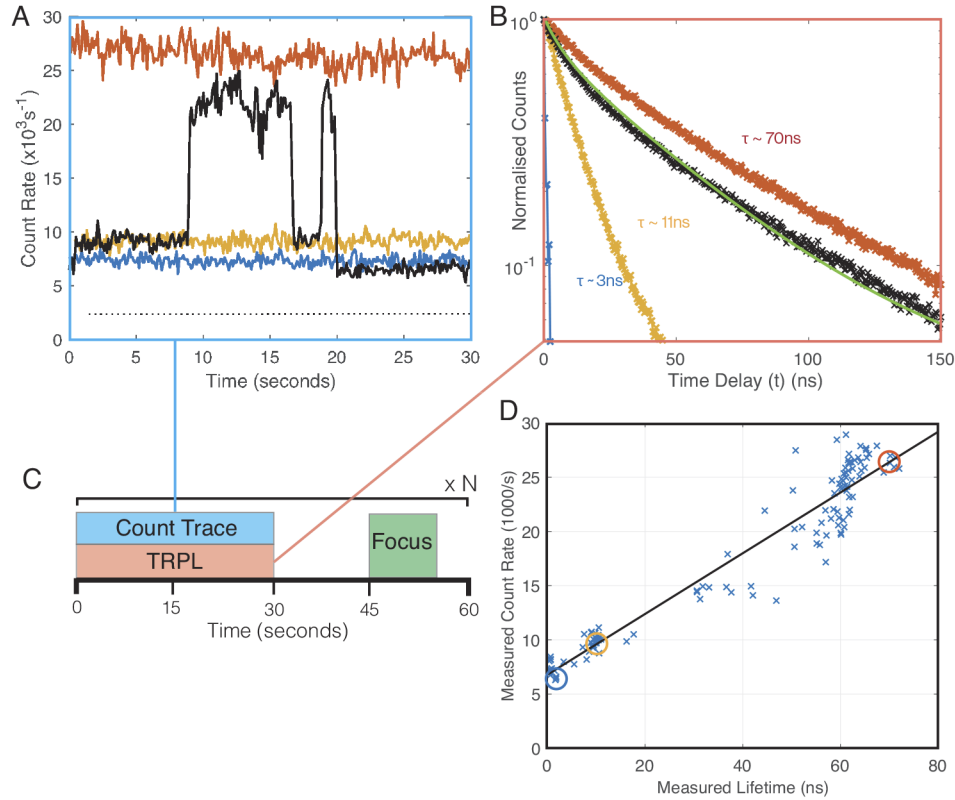


Fig. 6. Short term emission instability of NV2 following extended excitation. Simultaneous TRPL and count trace measurements taken. A: Four, of 184, 30 second traces from NV2. B: Measured TRPL during exposure period of traces in A. Predicted decay curve by integral sum of decay components from corresponding trace shown in green. C: Schematic of measurement cycle under 5 MHz excitation. D: Count rate level and corresponding fitted lifetime show linear correlation. Background count rate = 6786s^{-1} . gradient $(I - I_{bkg})/\tau_{rad} = 280.7$.

The NV fluorescence switches between brightness levels on a timescale of seconds. Where a TRPL measurement is taken with multiple brightness levels, a multi-exponential curve is observed. Where only one brightness level is observed (as with the three example traces in Fig. 6(A)), a mono-exponential TRPL curve is generated, with the decay lifetime changing with the brightness level. Figure 6(D) reveals a strong correlation between the measured brightness and measured lifetime throughout the recorded time duration, suggesting that the switching events are due to sudden changes in the non-radiative relaxation rate of the NV centre. Decay curves can therefore be predicted, given the count trace, by an integral sum of the decay components. One example, from the black trace in Fig. 6(A) is included in Fig. 6(B).

3.3. Long-term stability of NV centres in 100 nm nanodiamonds

Nanodiamonds with a nominal diameter of 100 nm generally demonstrated greater stability compared to those from the smaller sample. Once again, a range of emission stability was observed - attributable to varying positions of the NV centres relative to the nanodiamond surface. In the most stable examples (2 of 6), less than 10% variation in measured lifetime was observed

over an extended period of 60 hours, with mono-exponential decay curves throughout. Brightness variations are attributable to defocus of the excitation laser. The measured lifetime and 5 MHz rate for these most stable NV centres is included in Fig. 7. That this degree of stability was observed twice in such a small sample size suggests that it is not exceptional behaviour.

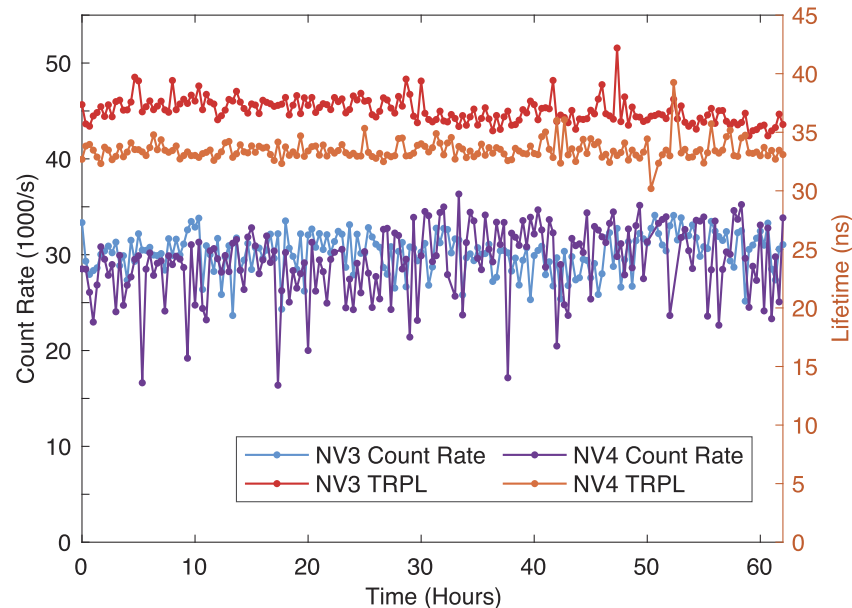


Fig. 7. Stability in fluorescence decay dynamics for two 100 nm nanodiamond hosted NV centres. NV3 and NV4, demonstrate less than 10% variation in measured lifetime and stable emission over 62.5 hours of 80 MHz pulsed excitation.

4. Discussion

Observed lifetime distributions are in agreement with those reported elsewhere [12], with much shorter lifetimes than predicted assuming unity quantum efficiency. ALT results show two trends in emission over time in the smaller nanodiamonds over a prolonged period of pulsed excitation. First a slow, progressive increase in the radiative lifetime and second a reductions in quantum efficiency and emission stability. An increased radiative lifetime is most likely to result from the gradual reduction of the optical density of states (DOS) in the NV environment which we propose is due to host nanodiamond size reduction. The optical DOS is independent of the location of the NV centre within the nanodiamond, as it is defined by a length scale on the order of the optical wavelength [9–12]. We propose the reduction in quantum efficiency is due to the laser-mediated introduction of discrete sites on or near the nanodiamond surface, which introduce nonradiative relaxation pathways, the uptake of which is highly sensitive to the sites' proximity to the NV centre [13,15]. The effect of individual nonradiative sites being introduced or removed is observed as sudden changes in intensity and relaxation lifetime reported in Fig. 6, while the increasing proximity of these sites to the NV centre as the nanodiamond shrinks leads to the long term dimming and ultimately quenching of fluorescence as shown for NV1 in Fig. 4.

The mechanism responsible for the indicated size reduction is presently uncertain. The laser excitation power and frequency used in the experiments is well below the level at which heating is expected to oxidise the nanodiamond surface [22]. Work by Kononenko et al. on bulk diamond surface nano-fabrication has identified an oxygen-mediated nanoablation process under pulsed illumination [21,23–25]. They report a nanoablation rate, on a bulk diamond surface, of

approximately 10^{-4} nm/pulse with a 100 femtosecond pulse duration, a fluence of 600 mJ cm^{-2} , at a laser wavelength of 266 nm and approximately 5×10^{-6} with the same pulse duration, a fluence of 1300 mJ cm^{-2} , at a laser wavelength of 400 nm [21]. Their work suggests a power law dependence on laser fluence, with an exponent of 2 or 4 depending on the wavelength. Changes in emission we observe, suggest a size reduction rate of about 10^{-12} nm / pulse. This much slower rate, compared to the nanoablation work, could potentially follow a similar mechanism, despite lower fluence of about 11 mJ cm^{-2} , longer pulse duration of 100 picoseconds, longer wavelength of 532 nm and nanodiamond surfaces being different to those of bulk diamond. Future work, giving direct measurement of nanodiamond size, before and after a period of extended exposure from a pulsed laser with power, pulse duration and wavelength relevant to single photon source device use would be informative. However, limited NV centre electron-beam stability, and the high likelihood of host nanodiamond rotation during atomic force microscopy scans, limit the possibility of non-destructive host nanodiamond size measurement.

The lack of such a reduction in brightness and emission stability for the most stable NV centres in the larger nanodiamond sample, suggests the NV centres are sufficiently deep below the surface to be unaffected by the presence of non-radiative relaxation sites. Previous calculations show that the change in lifetime is non-linear with radius [11], such that the observed changes are expected to be smaller for the larger nanodiamonds for the same ablation amount. A prolonged experiment, investigating whether even the most stable NV centres in large nanodiamonds eventually deteriorate would be informative, albeit time consuming. Nevertheless, this result demonstrates the potential for stable near-unity quantum efficiency colour centre source, hosted within nanodiamond (with its associated advantages for collection efficiency and processing routes) exposed to a pulse number equivalent to 1000 hours at 5 MHz.

5. Conclusions

NV Centres within nanodiamond are among the most robust of fluorescent nanomaterials and are suitable for use as triggered single photon sources at room temperature, but the demands of practical device operation nevertheless require careful nanodiamond selection. We have shown that the prolonged, intense pulsed excitation required for such devices leads to progressive reductions in emission stability for NV centres within 50 nm host nanodiamonds due to a gradual reduction in nanodiamond size that we tentatively attribute to an oxygen-mediated nanoablation process.

Greater stability is observed in 100 nm nanodiamonds with examples of NV centres showing robust single photon emission over 10^{13} excitation cycles suggesting that NV centres deep inside these larger nanodiamonds are well suited to practical device applications.

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Disclosures

The authors declare no conflicts of interest.

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