# Time-dependent Mandel Q parameter analysis for a hexagonal boron nitride single photon source

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**Abstract:** The time-dependent Mandel Q parameter, Q(T), provides a measure of photon number variance for a light source as a function of integration time. Here, we use Q(T) to characterise single photon emission from a quantum emitter in hexagonal boron nitride (hBN). Under pulsed excitation a negative Q parameter was measured, indicating photon antibunching at an integration time of 100 ns. For larger integration times Q is positive and the photon statistics become super-Poissonian, and we show by comparison with a Monte Carlo simulation for a three-level emitter that this is consistent with the effect of a metastable shelving state. Looking towards technological applications for hBN single photon sources, we propose that O(T) provides valuable information on the intensity stability of single photon emission. This is useful in addition to the commonly used  $g^{(2)}(\tau)$  function for the complete characterisation of a hBN emitter.

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### 1. Introduction

Hexagonal boron nitride (hBN) is a two-dimensional wide bandgap semiconductor material which hosts atomic vacancy defects. These defects are now widely studied because they can produce bright single photon emission even at room temperature [1,2], with the possibility of being integrated into two-dimensional heterostructure devices [3,4]. Single photon sources based on hBN are beginning to be applied in quantum information technologies, for example in quantum key distribution (QKD) [5,6], fundamental tests of quantum theory [7], as well as integrating quantum emitters with optical cavities [8,9,10,11].

A key component of quantum optical technologies is the single photon on demand (SPoD) source: a source which can deliver deterministic single photon pulses at high count rates [12,13]. These sources may find applications in QKD [14], optical quantum computing, radiometry [15,16], or even in probing the single photon response of biological systems [17]. For a quantum emitter such as those in hBN to be used as a SPoD source, it must have a photon number distribution as close as possible to the single photon number state. In other words, both multiphoton and zero photon pulses must be suppressed so that the photon number variance is reduced. The range of timescales over which this small photon number variance is preserved is a measure of the intensity stability of the source. Quantifying this stability is our motivation to measure the time-dependent Mandel Q parameter for a quantum emitter in hBN.

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The time-dependent Mandel Q parameter is defined as the variance over mean minus one for the photon number N measured from a source during integration time T [18,13,19]:

$$Q(T) = \frac{\left\langle \Delta N^2 \right\rangle_T}{\left\langle N \right\rangle_T} - 1 \ . \tag{1}$$

 $\langle \ldots \rangle_T$  denotes averaging over the integration time T. The photon number for a coherent state follows a Poisson distribution, therefore  $\langle \Delta N^2 \rangle_T = \langle N \rangle_T$  and Q(T) = 0. The expected result for a single photon source is a negative Q parameter, which indicates the photon number distribution is narrower than a Poisson distribution and so is an indication of intensity squeezing. An ideal single photon source detected with no losses has Q = -1.

In previous works Q(T) has been used to characterise single photon emission from a single molecule triggered single photon source [13]. Other more varied applications outside quantum information technologies include the use of Q(T) for single molecule measurements, for example the detection of singlet oxygen or the fast recognition of single dye molecules [20,21]. We note that the abstracts in Refs. [22,23] mention observations of negative Q(T) on timescales of  $\sim 10$  ns for hBN emitters, therefore this topic demands a full study.

In this paper we will present the characterisation of a stable quantum emitter in hBN, focusing on the time-dependent Mandel Q parameter. With the help of data simulated by Monte Carlo methods, we show that the effect of the detector deadtime prevented us from observing antibunching in the Q parameter under CW excitation. However, for pulsed excitation with appropriate parameters, a negative Q parameter was found when integrating over one pulse period  $(100 \, \text{ns})$ . Finally, we discuss the use of Q(T) to complement other measures in the complete characterisation of photon statistics for hBN quantum emitters. While the absolute value of the Q parameter depends on the collection efficiency of single photons, we propose that the crossover time at which the photon statistics change from sub- to super-Poissonian is a robust and useful measure of the emitter's intensity stability.

#### 2. Methods and materials

#### 2.1. Experimental setup

Solvent-exfoliated multilayer hBN nanoflakes (Graphene Supermarket) were dropcast onto a  $5\times5$  mm Si/SiO<sub>2</sub> substrate, followed by hotplate annealing at  $500^{\circ}$ C to remove carbonaceous material. The quantum emitters were then activated by thermal annealing at  $850^{\circ}$ C under an Ar atmosphere.

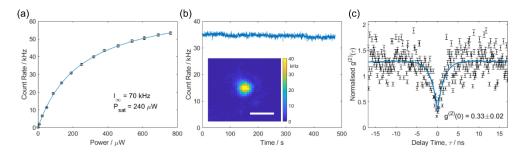
Samples prepared in this way were transferred to a custom-built confocal microscope in order to study photoluminescence (PL) from individual quantum emitters. The setup uses either continuous wave (CW) (Prometheus, Coherent Inc.) or picosecond pulsed (PicoQuant, pulse length 72 ps) lasers at 532 nm. The input beam is focused through a 100x (NA 0.9) objective onto the sample mounted on an XYZ piezo stage (PI Ltd.).

PL from the sample is collected through the same objective and separated from the pump beam by two dichroic mirrors (transmitting > 550 nm). The final stage of the setup is spectral filtering: first a long-pass interference filter transmitting > 568 nm (Semrock) removes the remaining 532 nm beam, then an angle tunable band-pass filter with a 20 nm bandwidth tunable from 560-630 nm selects the spectral region with the highest PL intensity. The output light is collected into a multimode fibre (50  $\mu$ m core diameter). To prevent misalignment with the fibre coupler when the tunable filter is rotated, a compensating plate was added after the filter which is calibrated to rotate in the opposite direction to the filter and compensate the beam walk-off from the optic axis.

A multimode fibre beamsplitter splits the output light onto two single photon avalanche diodes (SPADs, MPD), in the Hanbury Brown and Twiss (HBT) configuration. This experimental configuration was used for all the following measurements.

# 2.2. Single photon emitter characterisation

Individual quantum emitters were located by scanning the sample piezo stage to produce PL maps of the surface under CW excitation, see Fig. 1(b). All the following measurements were carried out on one emitter which was exceptionally stable. No blinking was observed for this emitter under any excitation power we used over hours of measurement. The blinking we refer to here is a transition between a bright and dark state of the emitter (common to the literature on hBN [24,2]), not the transition to a long-lived shelving state as the term blinking is used in Ref. [13]. Emitters showing fluorescence blinking have different transition rates when they are in their bright and dark states [24]; we expect our emitter to be free from this effect.



**Fig. 1.** (a) Photoluminescence (PL) count rate saturation curve for our hBN quantum emitter. The saturation power  $P_{sat}$  is 240  $\mu$ W. (b) PL count rate over time with 250  $\mu$ W CW excitation showing stable emission rate. The emitter showed no blinking events under any of the conditions studied; this is also shown by the small error bars in the saturation curve. Inset: PL map (photon count rate in kHz) of the quantum emitter. Scale bar: 1  $\mu$ m. (c) Second order correlation histogram for 250  $\mu$ W CW excitation. A two-exponential fit was applied to obtain  $g^{(2)}(0) = 0.33 \pm 0.02$ , confirming single photon emission.

The count rate saturation with increasing pump power is shown in Fig. 1(a). Fitting the following function to the saturation curve gave a saturation power of  $P_{sat} = 240 \,\mu\text{W}$  [24]:

$$I = \frac{I_{\infty}P}{P + P_{sat}} + bP + c \ . \tag{2}$$

I: count rate,  $I_{\infty}$ : max. count rate, b: power dependent background, and c: constant background. The beam diameter at the sample surface was measured to be approximately 1  $\mu$ m. Single photon emission was confirmed by measuring the second order correlation function  $g^{(2)}(\tau)$  with a value of  $g^{(2)}(0) = 0.33 \pm 0.02$  for CW excitation at 250  $\mu$ W, Fig. 1(c). A two exponential fit was applied with antibunching/bunching timescales  $\tau_1$ ,  $\tau_2$  to obtain the value of  $g^{(2)}(0)$  (as in [1], except that we use both antibunching and bunching parameters A, B to account for non-zero values of  $g^{(2)}(0)$ ):

$$g^{(2)}(\tau) = 1 - (1+A) \exp\left(-\frac{|\tau|}{\tau_1}\right) + B \exp\left(-\frac{|\tau|}{\tau_2}\right) . \tag{3}$$

The effect of uncorrelated background counts (assumed to be Poisson distributed) was removed for the CW measurements using the signal to background ratio SBR estimated from the PL intensity map of the emitter. For the SBR, from which we define  $\sigma = SBR/(1 + SBR)$  (in Fig. 1(c),  $\sigma = 0.987$ ), the correction is [24,25]:

$$g_{corrected}^{(2)}(\tau) = \frac{g_{raw}^{(2)}(\tau) + \sigma^2 - 1}{\sigma^2}$$
 (4)

To produce the  $g^{(2)}(\tau)$  histogram, photon arrival times on the two SPADs were recorded using a photon counting module (Time Controller, ID Quantique). The time resolution of our setup is limited by the SPAD timing jitter, nominally 50 ps.

# 2.3. Mandel Q parameter calculation

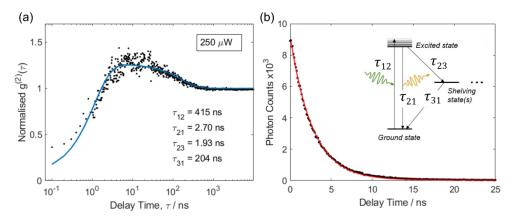
To measure the intensity stability of our emitter, the time-dependent Mandel Q parameter Q(T) was calculated according to Eq. (1). This was calculated using the same raw photon arrival times, or timestamps, used for  $g^{(2)}(\tau)$  measurements. Timestamps were collected in acquisition times of 100 s for all measurements.

To calculate Q(T), timestamps from both detectors were combined into one array, sorted, and split into K time windows of length T using a MATLAB script, included in Code 1 ([26]). Unless stated otherwise,  $K=10^8$  time windows were used (or the maximum allowed by the 100 s acquisition time for a given T) to calculate the variance and mean of the photon number per window  $\langle \Delta N^2 \rangle_T$  and  $\langle N \rangle_T$ , and hence Q(T). For pulsed excitation the T values were all multiples of the pulse repetition rate. Error bars were calculated by repeating the analysis over many acquisitions.

This analysis was carried out on timestamps from both CW (see Section 3.1) and pulsed (see Section 3.2) excitation. In addition to the procedure above, pulsed timestamps were also filtered using the trigger pulse from the laser, as in [13]. Only timestamps occurring within a window starting from the arrival of the laser pulse were kept. The optimum filter width was found to be 5 ns; this is discussed further in the Supplement 1.

# 2.4. Modelling hBN energy levels

In order to offer a comparison to experimental data for Q(T) and better understand the photodynamics of our hBN emitter, we simulated timestamp data using Monte Carlo methods. We used a three-level system as a simplified model for the emitter, defined by four transition lifetimes (see Fig. 2(b)). Although other works have found some emitters require four energy levels [24,27], we found that models with three and four energy levels fit our  $g^{(2)}(\tau)$  data equally well.



**Fig. 2.** (a) Fitting a three-level model to  $g^{(2)}$  data for CW excitation at 250 μW. The best fit transition lifetimes  $\tau_{ij}$  are shown; lifetimes for the 540 and 760 μW are in Table 1. (b) Radiative lifetime measurement made using pulsed excitation at 10 MHz and 24 μW mean power; data is shown for one 100 s acquisition. Fitting a single exponential to all 144 acquisitions of timestamp data gives a mean value of  $\tau_{21} = 2.7 \pm 0.1$  ns. Inset: Schematic of the three-level model for the emitter showing the transition lifetimes  $\tau_{ij}$  between levels.

Table 1. Fitting parameters for  $g^{(2)}(\tau)$  data

Power / µW	$ au_{12}$ / ns	$ au_{21}$ / ns	$ au_{23}$ / ns	$ au_{31}$ / ns	$\sigma$
250 (1.0 P <sub>sat</sub> )	415	2.70	1.93	204	0.962
$540 \ (2.3 \ P_{sat})$	189	2.83	1.85	195	0.722
$760 \ (3.2 \ P_{sat})$	135	2.86	1.81	125	0.811

Timestamps were simulated based on the method detailed in [25]. In each excitation cycle, the four lifetimes for each transition are drawn from exponential distributions to determine whether a photon is emitted (is the cycle radiative or non-radiative), and at what time. Losses are modelled by removing photon detections based on a binomial distribution, the detections are split 50:50 into two channels to model the beamsplitter in the HBT setup, afterpulses are added at random after some detections (see Supplement 1), and finally the detector deadtime is modelled by removing any detections less than the deadtime (80 ns for our SPADs) after the previous detection. The resulting timestamps are in an identical format to our experimental data, so the same data analysis for  $g^{(2)}(\tau)$  and O(T) was applied.

To run this simulation we needed to know appropriate transition lifetimes to set as mean values in the Monte Carlo model. We determined these parameters by fitting the three-level model to experimental  $g^{(2)}(\tau)$  data plotted on a log scale over the time interval 100 ps - 10  $\mu$ s by a procedure described in [24]. For a three-level system, the populations of the energy levels  $\rho_i(t)$  are described by a system of coupled differential equations:

$$\frac{d\rho_1(t)}{dt} = -k_{12}\rho_1(t) + k_{21}\rho_2(t) + k_{31}\rho_3(t) 
\frac{d\rho_2(t)}{dt} = k_{12}\rho_1(t) - k_{21}\rho_2(t) - k_{23}\rho_2(t) 
\frac{d\rho_3(t)}{dt} = k_{23}\rho_2(t) - k_{31}\rho_3(t).$$
(5)

The transition rates  $k_{ij}$  are used as fitting parameters for this model, which is related to the normalised  $g^{(2)}(\tau)$  by the solution to the differential equations for  $\rho_2(\tau)$ , with the initial conditions  $\rho_1(0) = 1$ ,  $\rho_{2,3}(0) = 0$  [28]:

$$g^{(2)}(\tau) = \frac{\rho_2(\tau)}{\lim_{t \to \infty} \rho_2(t)} \ . \tag{6}$$

This model will always have  $g^{(2)}(0) = 0$ , therefore to include the effect of the background counts we used the background correction Eq. (4) to include  $\sigma$  as a fitting parameter. The model was fitted to experimental data using the parameters  $k_{ij}$ , which are the reciprocals of the transition lifetimes  $\tau_{ij}$  represented in Fig. 2(b).

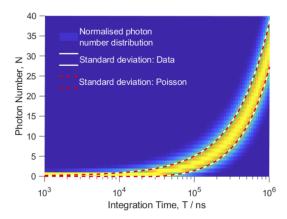
Additional constraints were needed to fit the four parameters  $\tau_{ij}$ : the radiative lifetime,  $\tau_{21}$ , was measured directly using pulsed excitation at 10 MHz and 24  $\mu$ W mean power. A single exponential fit to the lifetime curve in Fig. 2(b) gave  $\tau_{21} = 2.7 \pm 0.1$  ns. In addition, the excitation lifetime  $\tau_{12}$  was assumed to be linearly proportional to the power (as in [24]) and measurements at 250, 540 and 760  $\mu$ W were used to constrain  $\tau_{12}$ . The best fit parameters  $\tau_{ij}$  and  $\sigma$  at the three powers are given in Table 1, these were used as the initial parameters for the Monte Carlo simulated timestamps.

# 3. Results and discussion

# 3.1. Continuous-wave Mandel Q parameter

First, we examined the photon number distribution of our emitter for 100 s of timestamp data under CW excitation at  $250 \mu\text{W}$ , see Fig. 3. The single photon emission follows a distribution

very close to a Poisson distribution over integration times from 1  $\mu$ s to 1 ms. The photon number variance across these integration times is therefore almost the same as that of a coherent state. Since  $g^{(2)}(\tau)$  clearly shows antibunching over ns timescales (e.g. in Fig. 1(c), 2(a)), we would expect that as the integration time is decreased further the photon number variance will become significantly smaller than that of a Poisson distribution. The difference between the data and Poisson distribution variances is better visualized over several orders of magnitude in integration time using the Mandel Q parameter Q(T).

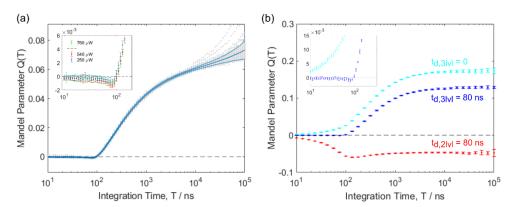


**Fig. 3.** Photon number distribution for CW emission under 250  $\mu$ W excitation. Along the y axis is a histogram of photon number per time bin, as a function of the time bin width or integration time, T. Each histogram height is normalised to one so the widths of the distributions can be compared. Curves indicate one standard deviation for the data (white), and a Poisson distribution with the same mean photon number (red). The data and Poisson deviations overlap almost exactly on this scale.

The Mandel Q parameter was calculated for CW excitation at three powers: 250, 540 and 760  $\mu$ W, see Fig. 4(a). We see that above approximately 100 ns the photon number distribution is wider than a Poisson distribution. Below this integration time there is a small negative value; the minimum is  $Q = -(1.4 \pm 0.3) \times 10^{-3}$  for 540  $\mu$ W, but Q(T) approaches zero at lower T. The measurement is limited at low integration times because the mean photon number per time bin tends to zero. At integration times  $\ll 1/I$ , where I is the emitter count rate, most time bins contain no photons. This is also illustrated by Fig. 3. For 250  $\mu$ W excitation,  $I = (34 \pm 3)$  kHz, therefore the average photon number is less than one below  $T = 29 \pm 3$   $\mu$ s.

Q(T) for simulated timestamp data can reproduce a similar form to the experimental data using the three-level Monte Carlo model described in Section 2.4. In Fig. 4(b) we compare the result for a two-level and three-level emitter, in which the transition lifetimes for the first two levels are kept the same (red and blue points, respectively). The transition lifetimes found in Table 1 were optimized to match the simulated Q(T) more closely to experiment and to have the same acquisition time as the experimental data. The final simulation parameters were:  $\tau_{12} = 205$  ns,  $\tau_{21} = 1.60$  ns,  $\tau_{23} = 1.40$  ns,  $\tau_{31} = 420$  ns, and  $\eta_{model} = 0.248$ . The two-level model uses only  $\tau_{12}$  and  $\tau_{21}$ , and  $\eta_{model}$  is the fraction of photons detected in the model.

Figure 4(b) illustrates that the positive Q(T) values are a consequence of adding a metastable shelving state. For a two-level emitter with only a ground and excited state (red points), the antibunched photon statistics and negative Q(T) are maintained as the integration time is increased. However, adding a shelving state (blue points) introduces a non-radiative decay path causing fluctuations in the single photon count rate. These become significant for integration times higher than the shelving state lifetime, which was  $\tau_{31} = 420$  ns in this case. Detector afterpulsing was



**Fig. 4.** (a) Experimental Mandel Q(T) for CW excitation at 250  $\mu$ W. Blue lines show mean values over 99 separate 100 s acquisition times, plus and minus one standard deviation. Inset: zoom-in showing small negative values for all three powers 250, 540 and 760  $\mu$ W. (b) Simulated CW Mandel Q(T) from Monte Carlo model. Blue (cyan) points were simulated using a three-level model with (without) modelling the 80 ns detector deadtime  $t_d$ . Red points are from a two-level model with the detector deadtime. Removing the deadtime effect makes all Q(T) values more positive, and for the three-level simulation removing the deadtime removes all negative values (inset). Comparing the two- and three-level simulations we can see that adding a metastable shelving state causes Q(T) to become positive at long integration times.

included in the simulated data in Fig. 4(b). and also causes Q(T) to become more bunched for T greater than the detector deadtime; see Supplement 1.

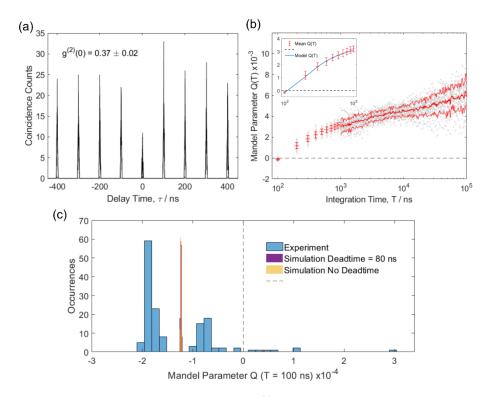
An important factor that must be considered is the detector deadtime. When the deadtime is artificially removed from the simulated timestamp data, Q(T) becomes positive for all integration times, i.e. the negative value disappears (cyan points in Fig. 4(b)). The deadtime introduces an artificial antibunching effect since there is a decreased probability of two photon detections within the deadtime period of around 80 ns. Therefore, the negative points in Fig. 4(a) are likely due to the detector deadtime only, especially considering the minimum value of Q(T) occurs near 80 ns. As discussed in Refs. [13,29], pulsed excitation with a repetition period longer than the deadtime must be used to avoid the deadtime artefact.

# 3.2. Pulsed Mandel Q parameter

For pulsed excitation, a 10 MHz repetition rate was used so that the repetition period of 100 ns was longer than the detector deadtime of 80 ns. A mean power of 24  $\mu$ W (pulse energy 2.4 pJ) produced a  $g^{(2)}(0)$  value of  $0.37 \pm 0.02$  (Fig. 5(a)) with a count rate of 2.8 kHz; increasing the power to 82  $\mu$ W and 160  $\mu$ W increased  $g^{(2)}(0)$  above 0.5 (see Supplement 1). The timestamps were filtered using a trigger pulse from the laser: only timestamps within 5 ns of the pulse start were used, i.e. approx.  $2\times$  the radiative lifetime. Four hours of timestamps at 24  $\mu$ W measured in 100 s acquisitions were used to produce the  $g^{(2)}(\tau)$  and Q(T) plots in Fig. 5.

We found a negative value at T = 100 ns of  $Q = -(1.4 \pm 0.8) \times 10^{-4}$ . As for the CW case, Q(T) is positive for integration times greater than 100 ns. This negative value is very small; the magnitude of Q(T) depends on the total detection efficiency of the measurement. The expected value for integration over one pulse period  $T_p$  is [29]:

$$Q(T_p) = \eta \left( \frac{g^{(2)}(0)}{2} - 1 \right) \tag{7}$$



**Fig. 5.** (a) Second order correlation function  $g^{(2)}(\tau)$  for 10 MHz pulsed excitation at 24 μW mean power.  $g^{(2)}(0) = 0.37 \pm 0.02$  was calculated using the relative area of the peak at zero delay time. Note that the timestamp data was filtered keeping only detections within 5 ns of the laser trigger pulse. (b) Mandel Q(T) for pulsed excitation at 24 μW showing positive Q(T) for integration times greater than 100 ns. Red lines show mean values +/-one standard deviation. Inset: fit to data ing Eq. (8). (c) Histogram of Q(T) values at T = 100 ns (integrating over one pulse period, blue bars). Values are compared over 144 acquisitions of 100 s each. The mean value is  $Q = -(1.4 \pm 0.8) \times 10^{-4}$ . The purple and yellow bars are Q(T) histograms from simulated timestamp data with comparable parameters, with/without modelling the detector deadtime. The distribution of Q(T) does not change when the simulated deadtime is removed, confirming that the detector deadtime has no effect on Q(T) when the pulse repetition period is greater than the deadtime.

where  $\eta$  is the total photon detection efficiency (i.e. including both the collection efficiency of photons and the detector efficiency). From our experimental values we can estimate  $\eta = (2 \pm 1) \times 10^{-4}$ . To measure a more negative value and a stronger signature of photon antibunching would require both a higher total detection efficiency and lower  $g^{(2)}(0)$ . We note that several works on hBN emitters have achieved low  $g^{(2)}(0)$  values (as low as 0.07) under pulsed excitation [3,5,6,30]. In our experiments we found a difficult trade-off in the pulsed excitation power between low  $g^{(2)}(0)$  improved by low power and achieving a sufficiently high count rate to make our measurements.

Simulated timestamp data was compared with or without the deadtime modelled in Fig. 5(c). The data were simulated with the method described in Section 2.4 using the parameters:  $\tau_{12} = 100$  ps,  $\tau_{21} = 2.70$  ns,  $\tau_{23} = 2.40$  ns,  $\tau_{31} = 420$  ns, and total detection efficiency  $\eta_{model} = 2.54 \times 10^{-3}$ .

There is no significant difference in Q(T) when the deadtime is removed for pulsed excitation at 10 MHz, and crucially, the negative value at  $T=100\,\mathrm{ns}$  remains. Therefore, we conclude that the experimental Q(T) data for 24  $\mu\mathrm{W}$  pulsed excitation shows evidence of antibunching at  $T=100\,\mathrm{ns}$ 

100 ns and bunching at longer integration times. As in the case of CW excitation, the bunching is attributed to the presence of a metastable shelving state.

The lifetime of the shelving state can be estimated by fitting the model from [29] to the pulsed excitation Q(T) data, expressed in terms of the shelving/deshelving lifetimes  $\tau_{23}$ ,  $\tau_{31}$ , the integration time  $T = k\tau_{rep}$  (k is an integer number of pulse repetition periods  $\tau_{rep}$ ), and the total detection efficiency  $\eta$ :

$$Q(k\tau_{rep}) = \eta \cdot \left[ \frac{\tau_{31}}{\tau_{23} + \tau_{31}} \left( \frac{2 - \beta}{\beta} - \frac{2(1 - \beta)}{k} \cdot \frac{1 - (1 - \beta)^k}{\beta^2} \right) - 1 \right]$$
(8)

where  $\beta = \tau_{rep}(1/\tau_{23} + 1/\tau_{31})$ .

A simplified form of this model was used to fit data from the single molecule triggered single photon source in [13] by assuming that  $\tau_{rep}/\tau_{23}\ll 1$ ;  $\tau_{rep}/\tau_{31}\ll 1$  is satisfied. Our hBN emitter does not meet these criteria:  $\tau_{rep}/\tau_{23}=0.67$  and  $\tau_{rep}/\tau_{31}=0.14$ , therefore we use the full expression Eq. (8) to fit to our Q(T) data (shown in Fig. 5(b) inset). This yields values of  $\tau_{23}=153$  ns and  $\tau_{31}=665$  ns.

We note that in comparison with the single molecule fluorophore in [13] (having  $\tau_{23} = 3.85$  ms and  $\tau_{31} = 250 \ \mu s$ ), the hBN emitter has a much shorter shelving state lifetime. This makes the window of observable negative Q(T) values, between a typical detector deadtime and the transition to Q(T) > 0, narrower and more difficult to observe.

The crossover time at which Q(T) = 0 is a useful metric as it gives a measure of the stability of a single photon source over long timescales. Our single photon emitter had a crossover between 100 and 200 ns. A perfect single photon source, such as an ideal two-level system, would have Q(T) < 0 for all integration times.

For certain applications of hBN single photon sources the photon number distribution will be critical, for example SPoD sources or in probing the single photon response of biological systems [17]. In these cases, we need to not only confirm single photon emission using  $g^{(2)}(\tau)$ , but also quantify the single photon variance over timescales which will be relevant to the chosen application. The Mandel Q parameter provides a measure of the photon variance, provided that artefacts due to the detector deadtime are avoided properly. While the absolute value of Q(T) is influenced by detection efficiency, the crossover time at which the photon statistics transform from sub- to super-Poissonian could be taken as a measure of long-term stability of photon antibunching from a single photon source.

Further improvements to hBN emitters for technological applications could look to improving the stability of emission over longer timescales. Currently, photon bunching seems to be inevitable over  $\sim 100\,\mathrm{ns}$  timescales due to the presence of a metastable shelving state. However, hBN quantum emitters with different chemical structures have significantly different energy level structures and shelving state lifetimes [31]. An emitter with a longer shelving state lifetime and higher preference for radiative decay should offer better single photon antibunching stability.

#### 4. Conclusion

We presented measurements of the time-dependent Mandel Q parameter Q(T) for a quantum emitter in hBN, which showed a very stable count rate and no fluorescence blinking. A comparison between experimental and simulated Q(T) for CW excitation demonstrated that artefacts due to the deadtime of the single photon detectors prevent us from measuring photon antibunching.

By using 10 MHz pulsed excitation instead, with the pulse repetition rate longer than the detector deadtime, a negative value of  $Q(T) = -(1.4 \pm 0.8) \times 10^{-4}$  was obtained at T = 100 ns. At longer integration times Q(T) becomes positive due to photon bunching caused by the metastable shelving state, an effect we could also demonstrate using the simulated data.

The transition from sub- to super-Poissonian photon statistics occurs between 100 and 200 ns, which is significantly shorter than the crossover time of a few µs for previous measurements on

a single fluorescent molecule [13]. There is therefore a narrow window between our detector deadtime of 80 ns and the crossover time in which O(T)<0 can be observed.

Monte Carlo simulations based on transition rates obtained by fitting to the long-timescale  $g^{(2)}(\tau)$  allowed us to simulate timestamps that were comparable to the specific hBN emitter being studied. This method proved useful for investigating the effects of the detector deadtime and metastable shelving state on Q(T) by allowing us to artificially remove these effects.

The Mandel Q parameter provides a measure of the intensity stability of a single photon source, in particular the crossover time indicates the integration time up to which photon antibunching is maintained. This is important for characterizing hBN single photon emitters towards applications in quantum information technologies such as SPoD sources.

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Data availability. See Code 1 [26] for MATLAB scripts used to calculate the time-dependent Mandel Q parameter.

**Supplemental document.** See Supplement 1 for supporting content.

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