

Coherence Properties of Electron-Beam-Activated Emitters in Hexagonal Boron Nitride Under Resonant Excitation

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Two-dimensional (2D) materials are becoming increasingly popular as a platform for studies of quantum phenomena and for the production of prototype quantum technologies. Quantum emitters in 2D materials can host two-level systems that can act as qubits for quantum information processing. Here, we characterize the behavior of position-controlled quantum emitters in hexagonal boron nitride at cryogenic temperatures. Over two dozen sites, we observe an ultranarrow distribution of the zero phonon line at approximately 436 nm, together with strong linearly polarized emission. We employ resonant excitation to characterize the emission lineshape and find spectral diffusion and phonon broadening contribute to linewidths in the range 1–2 GHz. Rabi oscillations are observed at a range of resonant excitation powers, and under 1- μ W excitation a coherent superposition is maintained up to 0.90 ns. Our results are promising for future employment of quantum emitters in *h*-BN for scalable quantum technologies.

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I. INTRODUCTION

The development of quantum technologies is expected to enable opportunities in enhanced sensing, secure communication, and quantum computation [1–4]. Solid-state quantum systems, in particular, have the potential to be efficiently integrated into scalable quantum technology devices [5], and platforms, such as diamond color centers, have already achieved milestones in this direction [6,7]. However, interest is growing in the use of layered two-dimensional (2D) materials, such as hexagonal boron nitride (*h*-BN), given their atomically thin nature. The exploration of defects in *h*-BN has shown this platform is capable of hosting both spin [8–13] and single-photon emitters (SPEs) [14–21], which could provide the basis for a spin-photon interface for use in quantum information distribution [22].

Despite progress, achieving desirable photonic capabilities from a SPE in *h*-BN continues to impose significant experimental challenges. For example, photon indistinguishability requires an emitter with a spectral linewidth

free of inhomogeneous broadening, but finding such a candidate has been nontrivial [23]. Moreover, *h*-BN hosts numerous SPEs with a broad range of emission wavelengths, and most defect fabrication methods are ineffective at generating SPEs consistently with a small variance in emission wavelength. An exception to this problem is a recently developed electron-beam irradiation technique for the generation of so-called blue emitters with an emission wavelength at approximately 436 nm [24–26]. The technique is site specific, it works reliably with high-quality *h*-BN crystals and the emitters can be produced en masse—all of which make the blue emitters appealing for studies of SPE spectral behavior and coherence properties. In addition, irradiation with keV electrons causes relatively little damage to the lattice, in comparison to competing site-specific methods, such as focused ion-beam irradiation [27,28], ultrashort laser pulses [29,30], and nanoindentation [31]. The condition of the lattice surrounding a defect has a significant influence on the emission lineshape and stability, and so minimally invasive defect fabrication is desirable for SPEs with favorable coherence properties.

In this work, we characterize the nonresonant photoluminescence properties of the blue emitter at cryogenic temperatures. Resonant excitation is then used to

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characterize the emission lineshape, the degree of spectral diffusion, and linewidth broadening versus temperature. Most significantly, we demonstrate coherence of optical transitions through the observation of Rabi oscillations. This result is a key requisite capability for these blue emitters to be considered as a viable platform for the development of scalable quantum technologies in *h*-BN.

II. RESULTS

Spectral instability, such as spectral diffusion and blinking of quantum emitters in *h*-BN are known issues hindering practical application of such emitters [32,33]. The recently discovered blue emitter in *h*-BN, generated by electron-beam irradiation, was shown to have a stable emission intensity at around 436 nm. To investigate the emission properties of these emitters in detail, a flake of *h*-BN hosting blue emitters created using electron-beam irradiation is mounted in a cryostat and cooled to 5 K. The fabrication process of these emitters [26] is explained in Sec. IV. Three arrays of three-by-four sites are patterned on a single flake for this study. To reveal the locations of the emitters, a confocal photoluminescence (PL) scan is performed over a $10 \times 10 \mu\text{m}^2$ area containing each array. The arrays are here termed matrix A (MA), matrix B (MB), and matrix C (MC). These confocal scans are performed using a 405-nm laser operating at 1 mW, with collection filtered using a $436 \text{ nm} \pm 3 \text{ nm}$ band-pass filter. Figure 1(a) shows the confocal PL scan over MA, and confocal scans of the other arrays are presented in the Supplemental Material [34]. Measurements of the high-resolution PL emission spectrum from each site in MA are shown collectively in Fig. 1(b). Each spectrum is normalized and offset vertically for clarity. The twin emission maxima evident in the spectrum plotted second from the top is most likely from the presence of multiple emitters at this site. Time series of PL spectra revealed no discernable temporal variation in intensity (blinking) or peak wavelength (spectral diffusion), as seen in Fig. S1(d) within the Supplemental Material [34]. A broadband PL spectrum collected using a 430-nm-long pass filter is shown in Fig. S5 within the Supplemental Material [34]. The dominant zero-phonon-line (ZPL) peak in the spectrum is accompanied by distinct low-energy and high-energy phonon sidebands (PSBs), which we attribute to longitudinal acoustic and longitudinal optical modes, respectively [35].

PL spectra are also measured for sites located in MB and MC, and the distribution of peak emission wavelengths is shown in Fig. 1(c). Across the 46 sites measured, the average ZPL wavelength is $435.98 \text{ nm} \pm 0.21 \text{ nm}$. The small variability in emission wavelength reflects a high degree of consistency in the spectral properties of this class of emitter, which we attribute to preferential fabrication of a specific defect structure by the electron beam, and a high purity and crystallinity of the *h*-BN sample grown by

the high-pressure high-temperature (HPHT) method. We note that this narrow distribution of emission wavelength is uncommon in other classes of emitters in *h*-BN [9,36].

Previous work has speculated on the nature of the SPE formation under electron-beam irradiation and the resulting defect structure, which may feature substitutional, vacancy, or interstitial atoms [24–26]. Structural insight may be gained indirectly through measurements of the emission dipole direction and polarizability. With this in mind, polarization measurements are performed at each of the available sites with the aim of observing a potential correlation between the most common polarization directions and the orientation of the underlying *h*-BN crystal lattice, which has threefold rotational symmetry.

To study the emission polarization, emitters are illuminated with a circularly polarized laser at 405 nm. The polar plot in Fig. 1(e) shows a polarization measurement typical of the sites in MA. Further polar plots in Fig. S2(b) within the Supplemental Material [34] show the results of polarization measurements performed at each site in MA. The data are normalized and fit with a cosine function:

$$p = \alpha \cos^2(\theta - \theta_0) + \beta.$$

The polarizability is defined as the difference between the maximum and minimum values of the fitted function. Polarization measurements are performed on all 46 sites located on the same flake, and the histogram in Fig. 1(d) shows the distribution of polarization angles observed. The bin size is 5° , and the data are segregated according to an arbitrary polarizability threshold of $p = 0.85$. Although there is clustering for angles corresponding to a high degree of polarizability, there is no clear correlation between the lattice symmetry angle and the clustering of polarization directions, consistent with the result from Fournier *et al.* [25]. The polarizability threshold is used in an attempt to filter out the low polarizability caused by the presence of multiple emitters in one site. There appears to be no strong correlation between the polarizability and the single photon purity as determined by the second-order correlation function (see Fig. S2 within the Supplemental Material [34]). That is, sites with relatively low polarizability are also found to be single-photon emitters.

To explore spectral properties of these emitters beyond the resolution limit of the PL spectra in Fig. 1(b), resonant photoluminescence excitation (PLE) measurements are performed. A Ti : Sap laser operating at $1 \mu\text{W}$ is used to scan across the ZPL of an emitter, with the resulting fluorescence collected from the PSB. In Fig. 2(a), the data sets show the fluorescence intensity from ten consecutive scans across the ZPL, with each scan offset vertically for clarity. The data from each scan are fit with a Lorentzian function, and for 50 consecutive scans the average linewidth is 1.73 GHz, taken by averaging the FWHM of each fitted

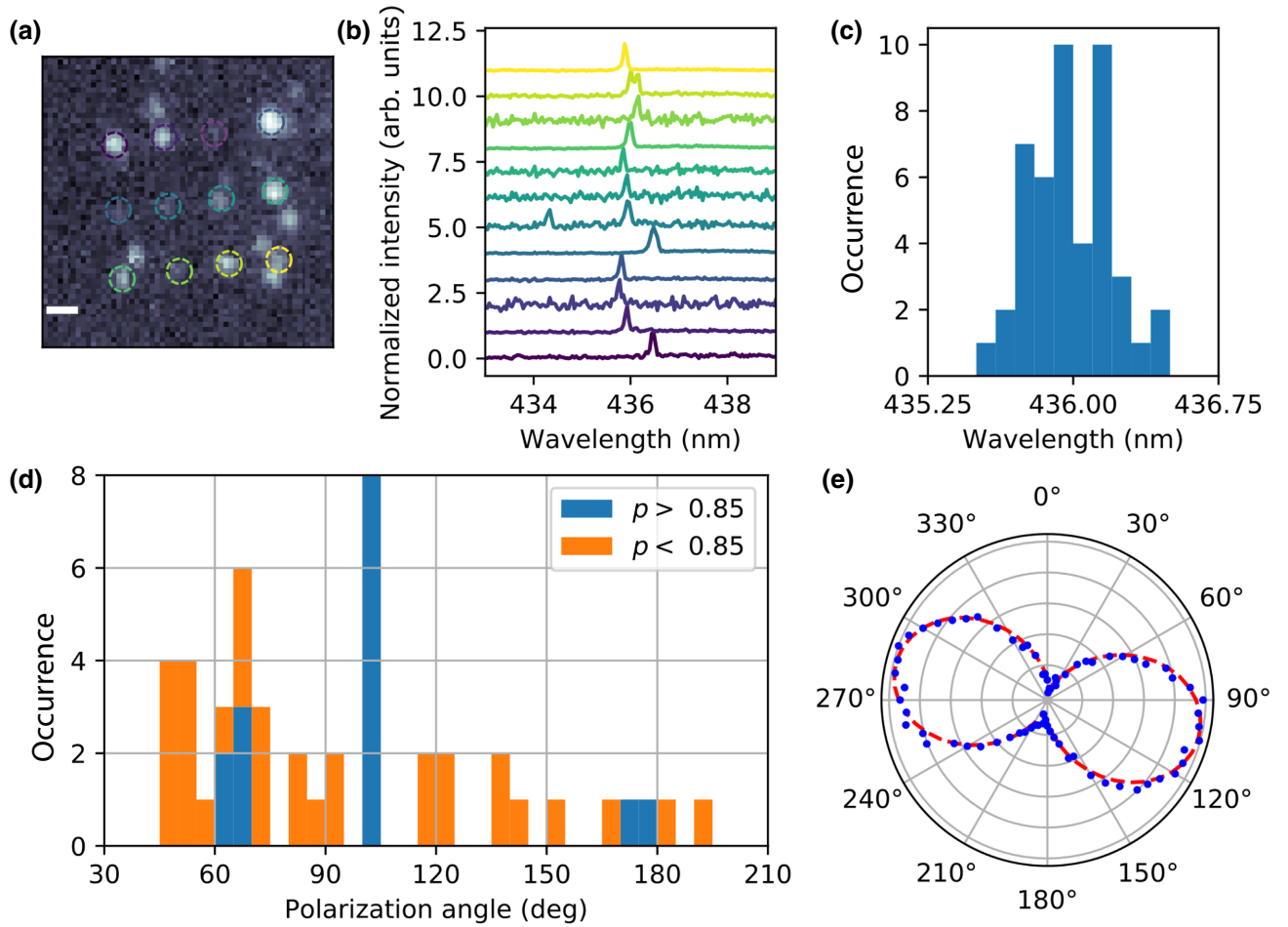


FIG. 1. Emission characterization. (a) Confocal PL scan of the 12 sites in matrix A (MA), one of three emitter arrays investigated in this work. Scale bar, 1 μm . (b) Normalized PL spectra measured at each site circled in (a). Each plot is vertically offset for clarity. (c) Histogram of ZPL wavelengths extracted from PL spectra across 46 emitter sites at three separate locations on the h -BN flake. The mean wavelength is $435.98 \text{ nm} \pm 0.21 \text{ nm}$. (d) Histogram of emission polarization angles measured from 46 sites, segregated by a polarizability threshold of 0.85. (e) A typical polarization measurement with data in blue and a sinusoidal fit in dashed red.

curve. The average ZPL is approximately 435.745 nm, or 688 017 GHz. The consecutive fits also indicate a slight degree of spectral diffusion, as shown by the frequency variation at which the peak intensity occurs. The set of ZPLs is approximately normally distributed, and fitting a Gaussian function to this distribution yields a FWHM of 0.91 GHz, representing an estimate of the extent of inhomogeneous broadening.

Because the magnitude of spectral diffusion is less than the average single-scan linewidth, the aggregate lineshape after many scans remains predominantly Lorentzian. This is shown in Fig. 2(b), where the average intensity over a given frequency interval for all 50 consecutive scans is plotted, together with both a Lorentzian fit and a Gaussian fit that, respectively, model homogeneous and inhomogeneous linewidth broadening. A Voigt function (i.e., a convolution of a Gaussian and a Lorentzian function) fit is also shown in Fig. 2(b). Each function yields a similar

linewidth, shown as inset values. However, an analysis of the residuals from each fit shows that the Voigt function gives the least error, and hence we conclude both homogeneous and inhomogeneous mechanisms are influencing the total linewidth. The Voigt fit allows the linewidths of the constituent functions to be extracted, and for these data the contributions to the convolution are $\Gamma_G = 1.50 \text{ GHz}$ for the Gaussian component, and $\Gamma_L = 1.06 \text{ GHz}$ for the Lorentzian component, which are close to the values obtained from the analysis of the 50 consecutive scans.

The emission linewidth in the absence of broadening processes is determined by the lifetime of the excited state, T_1 , through the uncertainty principle. Emitter lifetime is measured using a pulsed 405-nm laser operating at 100 μW with a pulse frequency of 20 MHz. From an exponential fit to the data, shown in Fig. 2(c), T_1 is determined to be $2.27 \text{ ns} \pm 0.02 \text{ ns}$, which is comparable to the lifetime of other emitter classes in h -BN [24–26,36]. Background

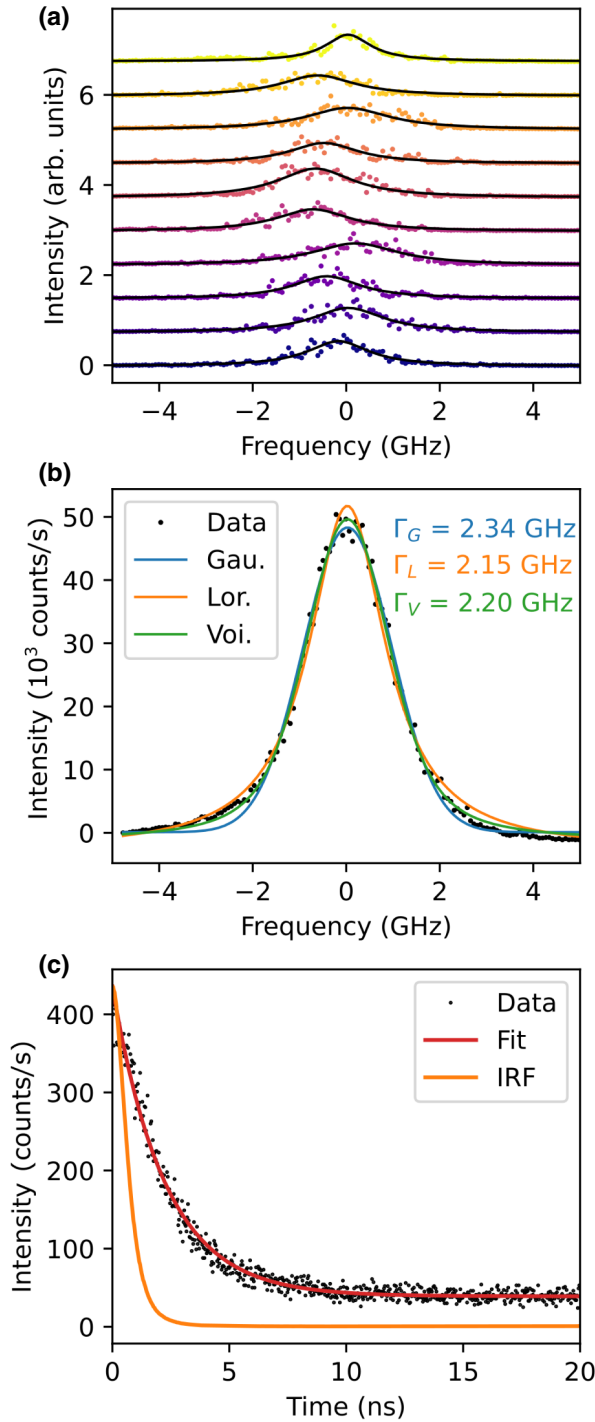


FIG. 2. Photoluminescence excitation. (a) Ten consecutive PLE scans across the ZPL of an emitter reveal some spectral wandering. Intensity data from each scan is fit with a Lorentzian function and offset vertically for clarity. (b) Average fluorescence intensity from 50 consecutive scans. These data are fit with a Gaussian function, a Lorentzian function, and a Voigt function, from which estimates of the linewidth Γ (FWHM) are extracted. (c) Lifetime measurement obtained using a pulsed laser. The exponential fit yields the excited state lifetime $T_1 = 2.27$ ns $\pm$ 0.02 ns. The orange curve is the instrument response function (IRF).

emission is measured at a location on the *h*-BN flake away from the emitter and used for a background corrected fit. A measurement of the instrument response function (IRF) is also shown in Fig. 2(c). The IRF data are normalized with respect to the maximum intensity observed over the emitter for ease of comparison. The lifetime measurement yields an estimate of $(2\pi T_1)^{-1} = 70$ MHz for the natural linewidth of this emitter. This value is much less than the linewidths obtained from the PLE scans above, confirming the presence of homogeneous and/or inhomogeneous broadening mechanisms.

We note that natural linewidths are reported for individual *h*-BN emitters fabricated by thermal annealing [37,38], however the distribution of ZPLs within this class of visible emitters is extremely broad, covering the range of 580 to 800 nm [39]. Such a wide variance impedes systematic analyses of defect structure and species and limits the ability to incorporate the emitter in quantum technology devices. In contrast, the blue emitters examined here employ a robust, site-specific electron-beam fabrication method to produce SPEs with highly consistent optical properties. This is advantageous and will accelerate advancements in studies of *h*-BN emitters for quantum applications.

The lineshape observed from PLE scans became more regular with increasing excitation power. Several scans across the ZPL of an emitter are performed for powers ranging from 0.04 to 2.4 μ W. Figure 3(a) shows the PLE data collected at each excitation power, with data sets for increasing power offset vertically. For each data set, the measured emission intensity is fit with a Voigt function. The same analysis undergone above indicates that this function is a better model for the intensity distribution than either the Gaussian or Lorentzian functions. The Voigt fit allows a quantification of the maximum intensity and linewidth of the ZPL, and in Fig. 3(b) the maximum intensity is plotted for each excitation power. These maxima display saturation behavior for increasing power and can be adequately described by the function,

$$I = \frac{I_\infty P}{P + P_0},$$

where I is the emission intensity, I_∞ is the maximum obtainable intensity, P is the excitation power, and P_0 is the saturation power. For this emitter, a saturation power of 3.14 μ W is extracted from the fitted function. Increasing excitation power up to saturation does not appear to have a strong influence on the linewidth. Figure 3(c) shows the FWHM of each Voigt fit plotted in Fig. 3(a), with the average linewidth found to be 2.13 GHz.

To investigate the influence of temperature on the emission spectrum of the emitter, PLE scans are performed using 1- μ W laser power, over 5 min of data collection,

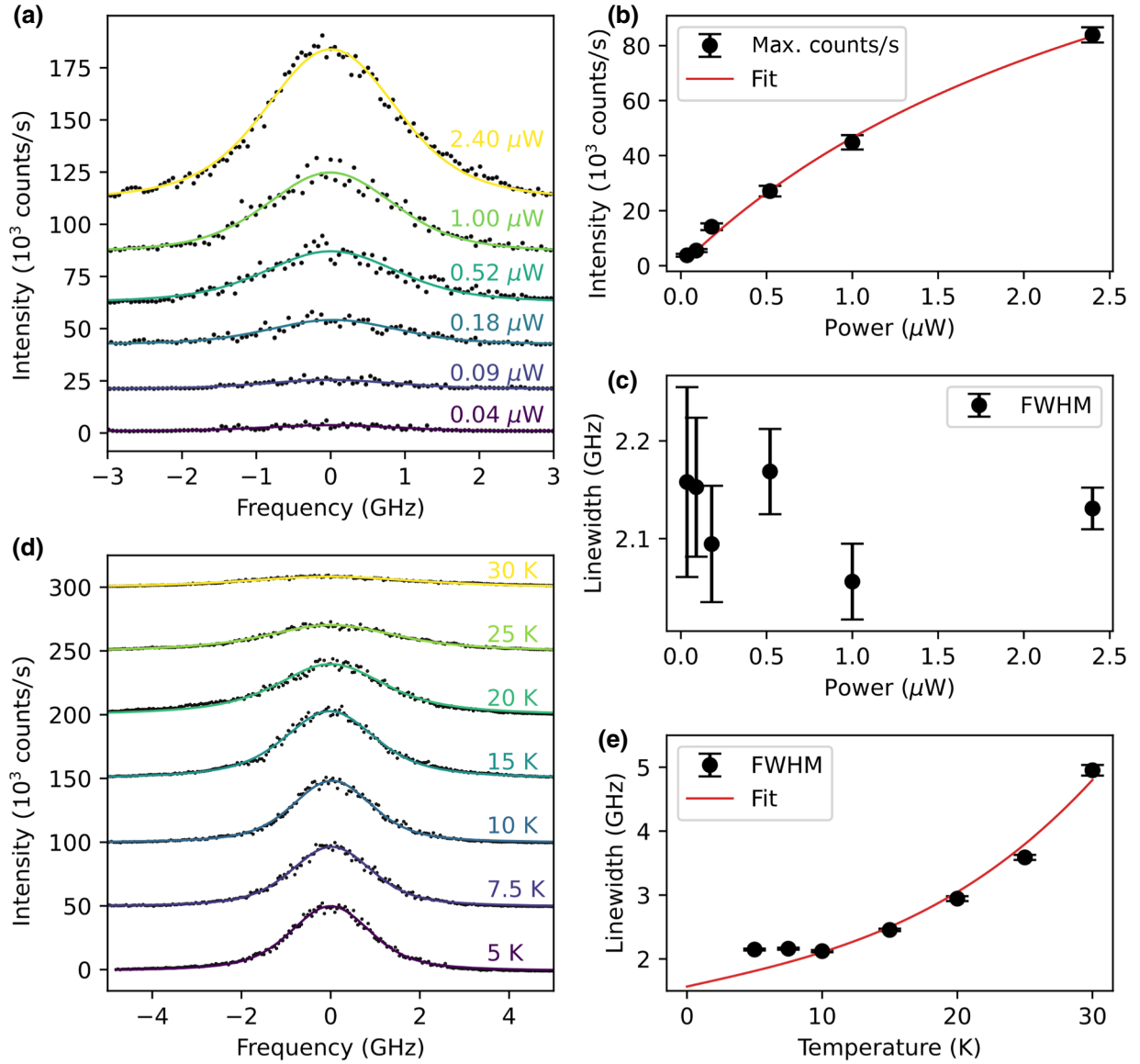


FIG. 3. Linewidth broadening. (a) PLE collected over a range of excitation powers. Each dataset is fit with a Voigt function and plotted with a vertical offset for clarity. (b) The maximum value of each Voigt function in (a) is plotted against the excitation power, and fitted with a power-intensity function to extract the saturation power 3.14 μW. (c) The full width at half maximum of each fitted function in (a) is plotted against the excitation power. (d) PLE collected at a range of temperatures, using an excitation power of 1 μW. Each dataset is fit with a Voigt function and plotted with a vertical offset for clarity. (e) The FWHM of each fitted function in (d) is plotted against the temperature and fit using a cubic function.

for a range of temperatures between 5 and 30 K. The line-shape is observed to widen in proportion to temperature, such that above 30 K the width had exceeded the maximum scanning interval of the laser, hence no data are collected past 30 K. The magnitude of inhomogeneous broadening is estimated by extracting the Gaussian component of the Voigt fit to the data at 5 K. This value of $\Gamma_G = 1.50$ GHz is then fixed for subsequent Voigt fits. The data sets and their Voigt fits are shown in Fig. 3(d), offset vertically with increasing temperature. Figure 3(e) shows the increasing FWHM together with a fitting function based on the

model [40–42]

$$\Gamma = \alpha T^3 + \beta T + \Gamma_0,$$

where Γ is the temperature-broadened linewidth, Γ_0 is the temperature-independent linewidth, and T is the temperature. The fit assumes that homogeneous broadening vanishes at 0 K, and hence only the natural linewidth of 70 MHz and inhomogeneous broadening of 1.50 GHz account for a fixed value of $\Gamma_0 = 1.57$ GHz. We attribute the discrepancy between the fit and the linewidth observed

at 5 and 7.5 K to additional broadening due to local heating [43,44].

When exciting at low power, the transition from the ground state to excited state occurs through spontaneous absorption of an incident photon from the excitation source. However, when the excitation laser is operating at high power, the emitter is driven to a coherent superposition of ground and excited states. Further, continuous high-power excitation modulates the probability amplitudes in a cyclic way, known as Rabi oscillation. The modulation of the excited-state probability is reflected in the bunching statistics of consecutive photons released by the emitter. Hence, Rabi oscillations are observable in measurements of $g^{(2)}$, the second-order correlation function. The Ti:Sap laser is used to resonantly excite an emitter at powers in the range of 0.1 to 2 μW . The collected emission is then directed to a fiber-based Hanbury-Brown and Twiss (HBT) setup. Figure 4(a) shows the second-order correlation at each power, using 32-ps bins and a total duration of 45 min. Each data set is fit with the exponential oscillation function [45]

$$g^{(2)}(\tau) = 1 - \exp(-\eta|\tau - \tau_0|) \left(\frac{\eta}{\Omega_R} \sin(\Omega_R|\tau - \tau_0|) + \cos(\Omega_R|\tau - \tau_0|) \right),$$

where $\eta = 3/(4T_1) + 1/(2T_2^*)$, T_1 is the excited state lifetime, T_2^* is the pure dephasing time, and $\Omega_R/2\pi$ is the Rabi frequency. The value of T_1 is fixed based on the lifetime measurement detailed above, while T_2^* and Ω_R are fitting parameters. The pure dephasing time decreases with increasing power, according to the fitted values shown inset above each data set in Fig. 4(a). The Rabi frequency should be directly proportional to the magnitude of the applied electric field, so that there is a square-root relation between incident laser intensity and observed oscillation frequency. Figure 4(b) shows that the Rabi frequency is well described by a linear fit to the square root of the excitation laser power, as expected. Similar measurements are performed at another emitter on the same sample, yielding comparable Rabi oscillations for resonant excitation in the tens of microwatts (see Fig. S4 within the Supplemental Material [34]). The increase in the value of $g^{(2)}(0)$ in proportion to power is attributed to increasing background emission. At a third site, no oscillations are observed, likely due to local environmental variation, which causes rapid dephasing and loss of coherence.

For the case of 1- μW resonant excitation, the pure dephasing time obtained from the fit ($T_2^* = 1.13$ ns) gives a coherence time [46] of $T_2 = 0.90$ ns. This describes the coherence time for two consecutive photons and gives a $2T_1/T_2$ ratio of approximately 5. This is much greater than the ideal ratio of 1, suggesting that two-photon interference

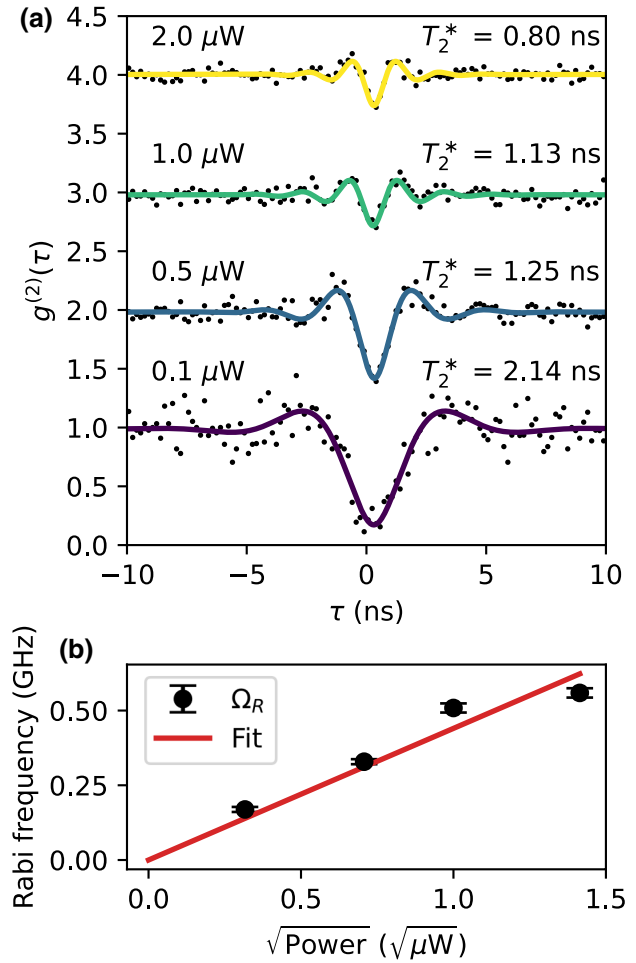


FIG. 4. Coherence properties. (a) Second-order correlation measurement collected over a range of excitation powers, and the fitted function demonstrating the presence of Rabi oscillations. Each plot is vertically offset for clarity. Values of T_2^* are extracted from the fit. (b) The Rabi frequency extracted from the fit in (a) is plotted against the square root of the excitation power, and a linear function is used to fit the data.

performed on this current system would yield far from perfect indistinguishability [47]. The coherence time is likely reduced further under the influence of phonon interactions and spectral diffusion that cause linewidth broadening [48], which would be relevant for the interference of photons with significant temporal separation. Nevertheless, Reimer *et al.* [44] note that Fourier-limited photons are not essential for observing indistinguishability, so long as the instrument response time is much shorter than the coherence time. This leaves the door open for future two-photon interference experiments with these blue emitters, which would be served in the meantime by complementary experiments aiming to quantify and control the rate of spectral diffusion [49,50].

III. CONCLUSION

This work represents an in-depth cryogenic study of a particularly promising *h*-BN defect. The blue emitters can be reliably fabricated to the extent that we are able to gather statistics on the spectral properties of several dozen emitters. In particular, we are able to achieve Rabi oscillations in autocorrelation functions measured from two different emitters. Power and temperature broadening measurements indicate that the linewidth is predominantly influenced by phonon-assisted recombination, although the relative contribution from spectral diffusion is less clear. Furthermore, lack of knowledge of the energy-level structure for this class of emitter invokes interest in the role of possible metastable states, and future work should consider the power dependence of the second-order correlation function to explore this feature. The observation of Rabi oscillations demonstrates that the defect associated with this blue emitter is capable of supporting a coherent superposition state, which further enhances the appeal of *h*-BN as a platform for quantum technologies.

IV. METHODS

A. Sample preparation

Bulk *h*-BN grown using the HPHT method is used to exfoliate a thin *h*-BN flake onto a SiO₂/Si substrate, followed by an annealing treatment in N₂ at 1000 °C. An isolated flake on the sample is then subject to electron-beam irradiation using a 10-kV beam, 1-nA current, and 2-s irradiation time per spot, resulting in the controlled formation of fluorescent defects at chosen sites on the flake. A total of three arrays of 3 × 4 defect sites are patterned onto the flake. The sample is then cooled to 5 K using a closed-loop cryostat. Further details on emitter creation are available in Ref. [26]. Further details on *h*-BN crystal purity are available in Ref. [51].

B. Photoluminescence experiments

PL spectra are measured using an Andor Spectrometer with 1800 lines/mm grating, integrating for 5 s under 1 mW of 405-nm laser excitation. To measure the emission polarization from each site the sample is illuminated with a circularly polarized 1-mW 405-nm laser. The fluorescence from each emitter site is directed through a rotatable (1/2)λ-wave plate (HWP) and onto a polarizing beam splitter. PL counts are then measured as the HWP is rotated through 180°. The lifetime measurement is performed using a pulsed 405-nm laser operating at 100 μW with a pulse frequency of 20 MHz. Emission is also measured at a location on the *h*-BN flake away from the emitter in order to perform background correction. For all PLE experiments, resonant excitation is performed with a frequency doubled Ti:sapphire scanning laser (M Squared) and photons are collected from the PSB. Data are collected

over 25-ms integration time. Scanning is performed at a rate of 1 GHz/s unless otherwise specified. Time series of PLE spectra are taken over 280 s at 1 μW. Second-order correlation measurements are performed using APDs and a time-tagging device (Swabian).

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The authors declare no conflicts of interest.

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