

Lifetime-Limited and Tunable Quantum Light Emission in h-BN via Electric Field Modulation

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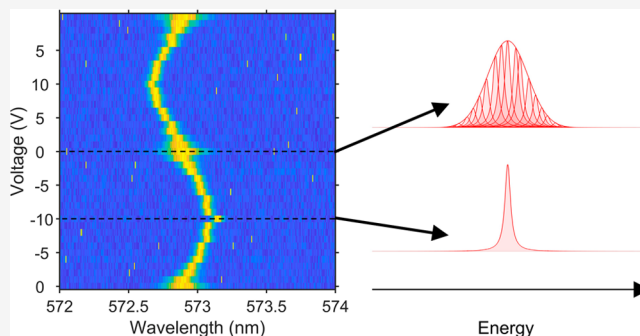
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ABSTRACT: Color-center-based single-photon emitters in hexagonal boron nitride (h-BN) have shown promising photophysical properties as sources for quantum light emission. Despite significant advances toward such a goal, achieving lifetime-limited quantum light emission in h-BN has proven to be challenging, primarily due to various broadening mechanisms, including spectral diffusion. Here, we propose and experimentally demonstrate suppression of spectral diffusion by applying an electrostatic field. We observe both Stark shift tuning of the resonant emission wavelength and emission line width reduction (down to 89 MHz) nearly to the homogeneously broadened lifetime limit. Finally, we find a cubic dependence of the line width with respect to temperature at the homogeneous broadening regime. Our results suggest that field tuning in electrostatically gated heterostructures is promising as an approach to control the emission characteristics of h-BN color centers, removing spectral diffusion and providing the energy tunability necessary for integrate of quantum light emission in nanophotonic architectures.

KEYWORDS: hexagonal boron nitride, single-photon emitter, electric field, lifetime-limited



Hexagonal boron nitride (h-BN) is a van der Waals material with a large band gap of ~ 6 eV. It has played a pivotal role in the development of 2D-material-based devices as an excellent gate dielectric and atomically smooth substrate.^{1,2} This material also hosts single-photon emitters in the visible range of the electromagnetic spectrum.^{3,4} Color centers in h-BN exhibit high stability at room temperature and above,^{5,6} as well as high Debye–Waller factors^{6–8} and potentially complete mechanical decoupling of the emitter from vibrations of the lattice.^{9,10} The atomistic picture of the origin of these emitters is still under debate,^{11–14} but the role of carbon in the formation of emitters in the visible range of the spectrum has been established.¹⁵ Optically driven magnetic resonance of ensemble^{16,17} and single¹⁸ emitters in h-BN shows that these spins can be magnetically addressed, useful for sensing and quantum computation applications.

Control of many quantum optical processes requires a source of spectrally indistinguishable photons.^{19,20} Thus, a narrow, ideally lifetime limited emission line width is therefore a crucial issue for such applications. While the color center point defects in h-BN are robust and bright sources of single photons even at room temperature, the emission spectrum is typically strongly affected by various broadening mechanisms. Previous studies have identified spectral diffusion as one of the major broadening mechanisms for h-BN single-photon sources, even at cryogenic temperatures.^{21–24} Spectral diffusion

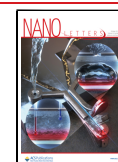
corresponds to rapid fluctuations in the energy of the emitter, which manifests itself as the rapid movement of narrow homogeneously broadened spectral lines. As a result, the observed line profile is a convolution of the homogeneously broadened spectral line and inhomogeneous spectral diffusion driven broadening ($\gamma = \gamma_{\text{hom}} + \gamma_{\text{inhom}}$). The homogeneously broadened line width depends on the lifetime and dephasing processes ($\gamma_{\text{hom}} = \frac{1}{2\pi T_1} + \frac{1}{\pi T_2^*}$). Here, T_1 is the decay lifetime of the emitter and T_2^* is the pure dephasing time scale which can result from spin–bath and/or emitter–phonon interactions. When the single-photon emission is purely lifetime limited, the inhomogeneous broadening is eliminated and the pure dephasing contribution to the homogeneous line width is zero, so the lifetime limit of the line width is $\frac{1}{2\pi T_1}$.

While the microscopic origin of spectral diffusion broadening is still under debate, there is a growing consensus that itinerant localized charges are a source of time-dependent

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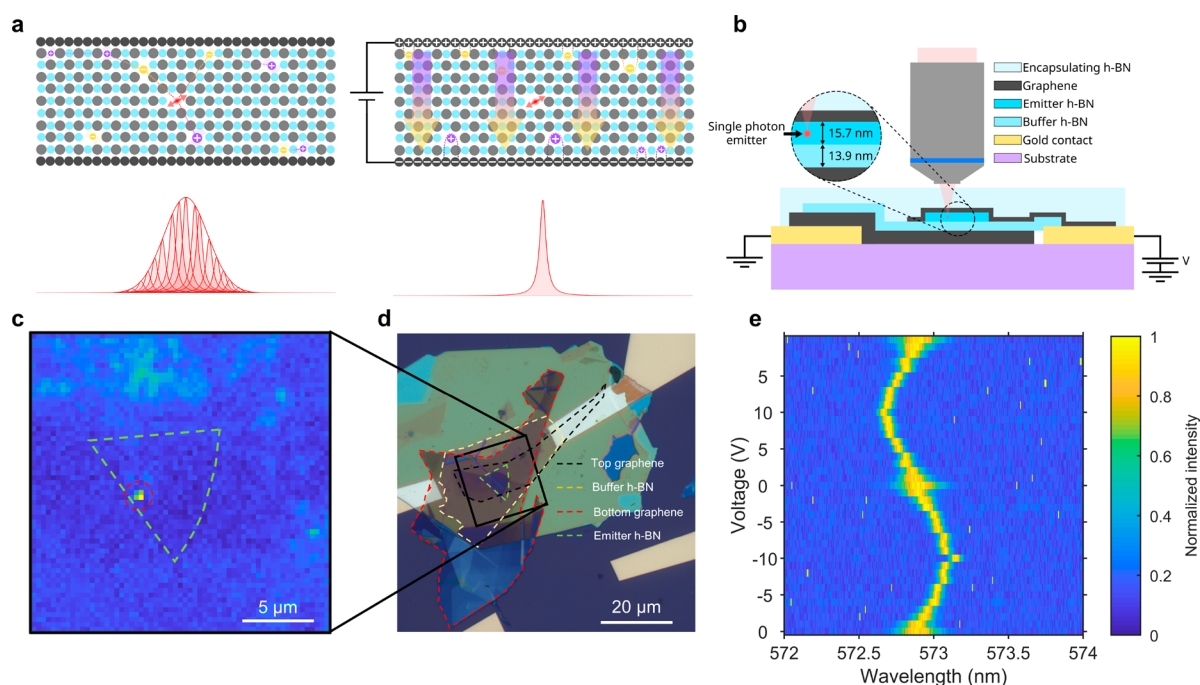


Figure 1. (a) Schematic of the effect of the electrostatic field on the single photon-emitter. The top panels show the emitter located in the h-BN crystal with and without the electric field, and the bottom panels show the emission spectra. An electric field locks charges located near the surface of h-BN at their place and reduces spectral diffusion. (b) Schematic of the device consisting of top and bottom graphene contacts, an h-BN-containing emitter (shown as the red dot), and buffer h-BN. (c) PL map of the region containing the emitter. Each pixel is the integrated spectra from 570 to 575 nm. (d). Microscope image of the device flake edges marked with dashed lines show the location of the top and bottom graphene and the buffer and emitter h-BN. (e) Normalized photoluminescence spectra of emitter ZPL at $T = 6.5$ K at voltages ranging from -10 V to 10 V.

electric fields in the emitter environment, which modulates the dipole overlap wave function leading to spectral diffusion. Photoionization of nearby impurities,^{23,25} charging and discharging of nearby charge traps,^{23,25} and the presence of mobile charged impurities²² are some potential sources of these localized charges. The time scale of spectral diffusion can range from microseconds²² to milliseconds^{23,24} to even seconds and minutes, depending on the dominant source of fluctuating environmental and stray fields.²⁶ Also, the dependence of spectral diffusion on pump power²⁴ supports the idea that the charges responsible for spectral diffusion can be photoactivated. So far, several studies have attempted different methods to suppress spectral diffusion in h-BN: for example, one study used anti-Stokes excitation to stabilize the spectral line,²⁶ but line width narrowing was not studied. Another study used transparent conducting substrates and reduced the inhomogeneous line width by 45%,²¹ but the resulting line width narrowing was still far from the lifetime limit. Another study showed that by combining a resonant excitation and lowering the excitation power, the effect of spectral diffusion was reduced.²⁴ In quantum dot single emitters, use of an electric field for charge depletion is established as a method to increase the indistinguishability of the emitted photons.²⁷

In this report, we present a new method to suppress spectral diffusion of an h-BN emitter and narrow the line width by nearly 2 orders of magnitude, approaching the lifetime limit of the emitter line width. To achieve this goal, we apply an out-of-plane electrostatic field to an emitter in h-BN and study its absorption spectra via photoluminescence excitation (PLE) spectroscopy.

For this experiment, an h-BN emitter with a zero-phonon line (ZPL) at 573 nm is studied. The emitter is induced in h-BN by the carbon-doping method explained in Methods; a

Voigt fit to the line shape reveals a ZPL line width of 0.90 meV at 6.5 K, which is higher than the average line width at cryogenic temperatures for emitters in thick exfoliated h-BN flakes,²¹ suggesting a significant broadening due to spectral diffusion.

The emitter is placed inside an all van der Waals device consisting of few-layer graphene electrodes and an additional buffer h-BN layer to prevent electrical shorting between the two electrodes as shown in Figure 1-b. A second-order intensity autocorrelation experiment shows $g^{(2)}(0) = 0.14$ indicating single-photon emission, while a time-resolved photoluminescence measurement reveals a decay lifetime of 3.36 ns, which corresponds to 47 MHz lifetime-limited line width as shown in Figure 2. The sample is cooled to 6.5 K, and the photoluminescence (PL) of the emitter is studied at voltages from -10 to 10 V at steps of 1 V, under 532 nm excitation. As can be seen in Figure 1e, the Stark effect produces a linear change in the energy of the ZPL, which suggests a nonzero out-of-plane dipole moment. The slope of the peak position as a function of voltage in both PL (Figure 1e and Figure S3) and PLE (Figure 3) measurements correspond to a value of 2.7 meV/(V/nm), considering the 29.6 nm thickness of h-BN.

In other studies, a Stark tunability of 43 meV/(V/nm)²⁸ for an in-plane electric field and 2.5–15 meV/(V/nm)²⁹ for an out-of-plane electric field are observed for h-BN emitters. Comparing with our result and assuming the dipole moments of different h-BN emitters to be comparable, we conclude that our emitter should be mainly in-plane with a small out-of-plane component. Polarization measurement of the emitter (Figure S2) also shows a linearly polarized emission, in accord with a small out-of-plane dipole.

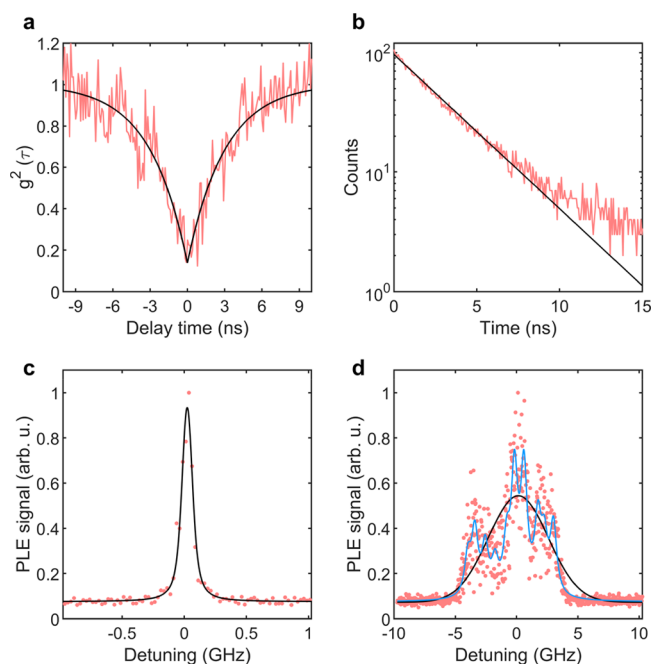


Figure 2. (a) Second-order autocorrelation function for photon emission. The fit has a value of 0.14 at $\tau = 0$. (b) Time-resolved photoluminescence of the emitter. The solid line corresponds to a single-exponential fit with a time constant of 3.36 ns; this value corresponds to a lifetime-limited broadening of 47 MHz. The deviation of data from the fit at $t > 10$ ns is a result of hitting the noise floor of the photodetector. (c) Normalized cryogenic ($T = 6.5$ K) PLE spectrum of the emitter at $V = 10$ V showing a line width of 89 MHz. (d) At $V = 0$ V PLE spectra show multiple peaks. The blue trace shows a fit with 10 Voigt functions with the same line shape as in (c), and the black trace shows a single Voigt fit to assign a fwhm value equal to 8.5 GHz.

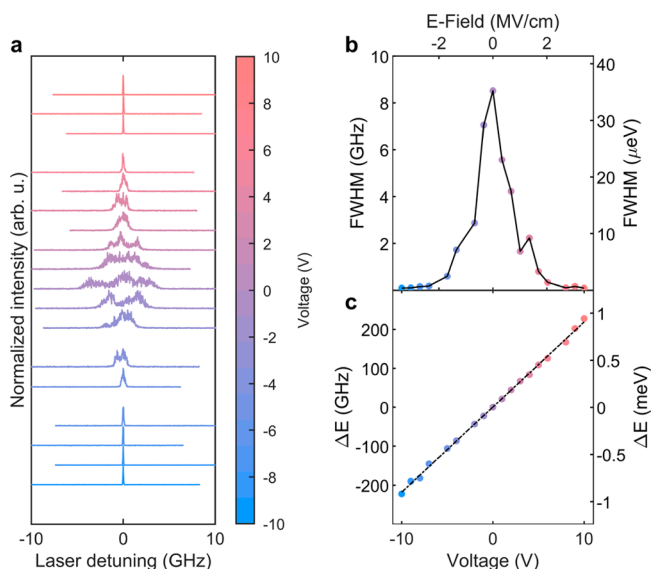


Figure 3. (a) PLE spectra of the emitter at $T = 6.5$ K for applied voltages ranging from -10 to 10 V. (b) fwhm of the peaks determined with a Voigt profile fit as a function of applied voltage. A minimum line width of 89 MHz is achieved. (c) Measured peak position of the emitter relative to its peak position at 0 V as a function of applied voltage.

More notably we see the line width narrowing as the voltage increases even in the nonresonant excitation regime (PL measurements). As the absolute value of voltage increases, the line width approaches the limits of spectrometer resolution equal to 0.2 meV. We note that as the emitter undergoes several cycles of voltage tuning the broadening at 0 V remains unchanged, suggesting negligible hysteresis in this process. This lack of hysteresis reveals that the charges responsible for spectral diffusion are not permanently removed/screened by the electric field. This supports the idea that the generation and recombination of the charges responsible for spectral diffusion is a dynamic process, without long-term charge trapping and that this dynamic process can be negated at a high enough electrostatic field. This effect might be caused by the excitation laser, charging the surrounding dark defects or mobilizing the charges in the surrounding of the emitter.

To further study the effect of electrostatic fields, without the limitations imposed by the resolution of grating-based spectroscopy and nonresonant excitation, we performed photoluminescence excitation spectroscopy (PLE) experiments using a continuous-wave, tunable, narrow line width laser source. In the PLE experiment, a tunable source of CW laser excitation is slowly scanned across the zero-phonon line and the intensity of the phonon sideband is measured as a function of laser detuning. As a result, this method can measure the absorption spectra of the ZPL with a resolution approaching the line width of the scanning laser source (~ 100 kHz). As shown in Figure 3, a two orders of magnitude reduction in line width is observed with the application of an electrostatic field, almost completely removing the inhomogeneous broadening effects at $V = 10$ V with a fitted line width of 89 MHz, as shown in Figure 2c. The difference between the lifetime limit of 47 MHz and the measured value for line width can have several possible explanations. One possible origin of this difference can be residual spectral diffusion; while the electric field has suppressed most of the effect of local charges that impose spectral diffusion, it is possible that some local charge dynamic still exists and the energy barriers to suppress these fluctuations are larger than the energy provided by the electric field. A more likely source of broadening can be dephasing processes. In such a case the observed line width will be limited by the total decoherence time scale consisting of lifetime and dephasing time scales. The source of this dephasing can be lattice vibrations, in which case a further reduction of the temperature can help to close the gap and achieve lifetime-limited emission.

The line shape of the emitter at $V = 0$ V (Figure 2d) and voltages near zero (Figure 3a) exhibits several peaks. This behavior is expected from spectral diffusion broadening, as it is caused by the spectral line jumping between several different energies. As PLE itself has a time scale comparable to the time for only a few jumps, the PLE spectrum does not show a single wide peak. The result of fitting with a sum of 10 Voigt functions to $V = 0$ V data can be seen in Figure 2d. This behavior makes it challenging to assign a value for full width at half-maximum (fwhm). So, we decided to assign a fwhm to these peaks by simply fitting a single Voigt function to the data and report the fwhm of the fitted peak. Even though this method does not capture the full complexity of the spectrum, it is effective in distinguishing between narrow and wide spectra and assigning a value for fwhm. The result is shown in Figure 3b. Atomic origins of this line width narrowing are not very clear, as the atomic structure of the emitters are still under

debate; we attribute the origin of the line width narrowing broadly to the changes in the local electronic charge fluctuations in the vicinity of the emitter. By applying an electric field, the charges in the h-BN are interacting with the DC electric field and this interaction can cause fewer fluctuations of local charges; this concept is depicted in Figure 1a.

It is worth noting that, in recent studies on the effect of electrostatic field on h-BN emitters,^{29,30} it has been observed that the emitter charge state can be modulated because of electrostatic doping, such that the emitter can switch between optically dark and bright states. However, we do not observe such an effect here. We suggest, that for emitters close to the h-BN surface, application of voltage induces electron tunneling through the h-BN barrier, thus populating and depopulating the emitter states in the near-surface region (within a few monolayers), effectively modulating the optical transitions from off to on. In the present work, a second h-BN layer was added to prevent electrical shorting between the two electrodes, and another consequence of this layer configuration is that it prevented electron tunneling, since the emitter is not in close contact with the electrodes. Our results suggest that the emitter is not within a tunnelling distance from the top electrode, implying also that it is closer to the bottom of the flake. The spatial distribution of h-BN emitters has not been very well studied, although several reports asserted that emitters are mostly located in the vicinity of edges of the flake.^{31,32} Also, one study on the axial location of emitters in h-BN has shown them to be close to the surface of a 300 nm thick flake of h-BN.^{33,34} Thus, it is possible that in most cases the emitters are either located near the surface or close to edges of the flake, which supports our understanding of both the recent studies cited, as well as this work. This phenomenon is possibly related to the microscopic effects of exfoliation and emitter generation.

The effect of temperature on line width has been discussed elsewhere,^{21,23,24} but the line width is usually convoluted with inhomogeneous broadening, so the pure effect of temperature on the emitter has been difficult to study independently from the effect of spectral diffusion. Since the electric field can suppress the contribution of the inhomogeneous broadening mechanisms on line width, we leveraged this opportunity to study the homogeneously broadened line width of the h-BN emitter and the effect of temperature on it. We observed a power law increase in the line width as the temperature increases, with the exponent $n = 3.02$ suggesting a T^3 dependence of homogeneous line width on temperature. This result is in accordance with some earlier studies on h-BN emitters.²³ However, some other studies have shown a linear dependence²¹ or a combination of linear and cubic, $AT+BT^3$,²⁴ dependence of line width on temperature. By carefully examining the results of these studies and comparing them with our results, we noticed that the study which used emitters with a strong spectral diffusion effect shows linear dependence,²¹ and the study which had a low-temperature line width of 1 GHz exhibits a $AT+BT^3$ dependence of line width on temperature, with the linear part being dominant below 20 K.²⁴ Comparing the results of these studies to our work, in which spectral diffusion is almost completely suppressed, leads us to conclude that spectral diffusion in h-BN is also activated by thermal fluctuations and exhibits a linear variation with temperature, but purely phononic line width broadening effects in h-BN result in a T^3 dependence of line width on

temperature (Figure 4). This is in contrast with diamond emitters, which show no dependence of spectral diffusion on temperature below 20K³⁵

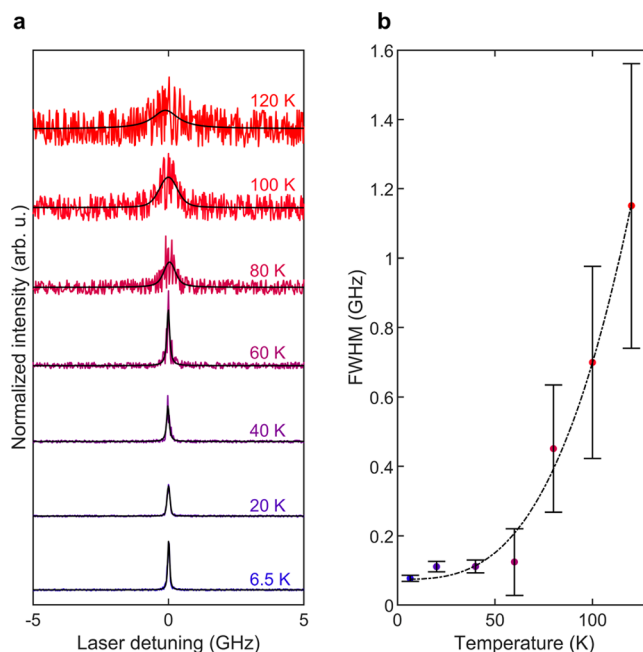


Figure 4. (a) PLE traces of the emitter at $V = 10$ V for various temperatures. (b) Fitted value of fwhm of the peaks as a function of temperature with a power-law fit ($\text{fwhm} = AT^n + C$) with a best-fit value of $n = 3.02 \pm 0.51$. Error bars correspond to 95% confidence intervals of the fitted values, and the minimum line width is 89 MHz at 6.5 K.

A T^3 dependence of line width on temperature is also observed in diamond nitrogen-vacancy (NV) centers.²⁵ In the case of NV centers, one possible explanation of the T^3 dependence is the fluctuating field created as phonons modulate the distance between the emitter and other defects and impurities, and as a result this type of dependence is mostly observed in samples with high disorder.³⁶ Our results suggest that this phenomenon is also present in h-BN. The surface of 2D flakes of h-BN is more prone to defects, and the emitters in thin layers of h-BN are always close to the surface, which can make this the dominant effect in h-BN. This is also in accordance with the large spectral diffusion at 0 V observed in our study, pointing to the existence of a very defective environment. This argument suggests that in an ideal case, in which spectral diffusion is fully suppressed and the defects to modulate local phonons are nonexistent, it is possible to have a minimal effect of temperature on line width as reported elsewhere.¹⁰

In summary, we have shown that an out-of-plane electrostatic field can suppress spectral diffusion while also Stark tuning the emission energy of hBN color center single-photon emitters. Our findings may pave the way for on-chip h-BN quantum communication technologies by removing the barrier to achieving lifetime-limited photons and potentially generating indistinguishable photons. The extra degree of freedom provided by Stark tuning is also desirable to couple the emitters to resonance modes of photonic cavities. Finally, we also studied the dependence of line width on temperature in

the lack of spectral diffusion and found it to be proportional to T^3 .

METHODS

Emitter Preparation. The emitter was prepared by annealing an h-BN crystal (HQ graphene) between two carbon tablets (MSE Supplies LLC, carbon sputtering target >99.99% pure) at 1200 °C for 10 h. Then, the h-BN crystal was exfoliated with Scotch tape onto a silicon oxide on silicon substrate. The exfoliated h-BN flake is 13.9 nm thick (see the Supporting Information), and an emitter is identified with a ZPL at 573 nm. This flake, containing the emitter is used in a device to produce a graphene-hBN-buffer hBN-graphene structure.

Device Fabrication. Graphene and buffer h-BN were directly exfoliated onto an SiO₂ (285 nm)/Si substrate and identified via their optical contrast. Substrates were cleaned via ultrasonication in acetone, isopropanol, and deionized water for 20 min each and then subjected to oxygen plasma at 300 mTorr and 70 W for 5 min. Exfoliation was done at 100 °C for a higher yield of flakes. All layers were assembled via the polymer-assisted hot pickup technique³⁷ (with polycarbonate/polydimethylsiloxane stamps). Flakes were picked up between temperatures of 40 and 70 °C and dropped at 180 °C. The polycarbonate was washed off in chloroform followed by isopropanol overnight (12 h). The assembly was done on a substrate of SiO₂ (285 nm)/Si with prepatterned electrodes/contacts of 5 nm Ti/95 nm Au fabricated with electron beam lithography followed by electron beam evaporation of the metal and subsequent liftoff in acetone and isopropanol. Finally, the device was wire-bonded onto a custom-made printed circuit board. The device was then transferred to an Attocube Attodry800 cryo microscope for optical characterization.

Optical Characterization. PL. We used a 532 nm CW laser (Coherent) as an excitation source for PL measurement at 100 μ W and used a spectrometer (Princeton Instruments) to measure the PL spectrum. The spectra acquisition time was 60 s.

g₂ and TRPL. A g₂ experiment was done at room temperature with 532 nm CW excitation at 100 μ W, the light was fiber-coupled to a fiber beam splitter (Thorlabs) connected to two avalanche photo diodes (Micro photon devices), and the single-photon events were registered by a PicoHarp300 electronics (Picoquant) instrument in TTTR mode. The signal was analyzed with a custom MATLAB script.

PLE. We used a tunable CW dye laser (Sirah Matisse); the signal from the emitter was passed through a 620 \pm 10 nm band-pass filter and was detected via an avalanche photodiode. The excitation wavelength was measured with a wavemeter (HighFinesseÅngstrom). PLE was performed at 10 nW power at a scan rate of 1 GHz/s.

ASSOCIATED CONTENT

Supporting Information

are available in the Supporting Information. The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.2c02163>.

Details of atomic force microscopy (AFM), dipole orientation and effect of the electric field on PL (PDF)

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Notes

The authors declare no competing financial interest.

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