Spectrally Resolved Photodynamics of Individual Emitters in Large-Area Monolayers of Hexagonal Boron Nitride

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†Supporting Information

ABSTRACT: Hexagonal boron nitride (h-BN) is a 2D, wide band gap semiconductor that has recently been shown to display bright room-temperature emission in the visible region, sparking immense interest in the material for use in quantum applications. In this work, we study highly crystalline, single atomic layers of chemical vapor deposition grown h-BN and find predominantly one type of emissive state. Using a multidimensional super-resolution fluorescence microscopy technique we simultaneously measure spatial position, intensity, and spectral properties of the emitters, as they are exposed to continuous wave illumination over minutes. As well as low emitter heterogeneity, we observe inhomogeneous broadening of emitter line-widths and power law dependency in fluorescence intermittency; this is strikingly similar to previous work on quantum dots. These results show that high control over h-BN growth and treatment can produce a narrow distribution of emitter type and that surface interactions heavily influence the photodynamics. Furthermore, we highlight the utility of spectrally resolved wide-field microscopy in the study of optically active excitations in atomically thin two-dimensional materials.

KEYWORDS: hexagonal boron nitride, super-resolution, semiconductor defects, single-molecule microscopy, 2D materials, multidimensional super-resolution, spectroscopy

Two-dimensional (2D) materials are rapidly emerging as promising for use in the areas of electronics,1 materials science,2 photovoltaic, light-emitting devices,3 and quantum emission.4–6 In the case of quantum technologies, the ideal solid-state system is sought that displays atomic-like states, which can be deterministically prepared and read-out optically.5,6 Hexagonal boron nitride (h-BN) has recently joined the small family of 2D materials that display single-photon emission.7 Previously, h-BN was widely used as a dielectric layer in graphene devices8,9 and was recently shown to display bright, visible single-photon emission at room temperature.10 The emission has been compared to that of nitrogen-vacancy (NV) centers in diamond, where the narrow-band visible emission is due to an atomic-scale point defect.10,11 The emissive defect state in h-BN is predicted to be either a boron vacancy (VB), nitrogen vacancy (VN), or an antisite complex (NBVN), in which a boron atom is replaced by a nitrogen atom with an absent neighboring nitrogen atom.7 Consequently, the
past few years have seen work on single-photon emitters in h-BN accelerate, but progress toward device integration has been hindered by the high variation in the frequency of the emission.\textsuperscript{12−14}

An explanation for the wide range of emission energies reported is the use of heterogeneous h-BN samples. So far, the vast majority of optical studies of h-BN have been carried out on multilayer samples without a well-defined sample thickness or morphology, namely, liquid\textsuperscript{7,12,13} or mechanically exfoliated\textsuperscript{15−17} h-BN, that are likely to contain a wider range of structural defects than chemical vapor deposition (CVD)-grown monolayer h-BN, due to the greater heterogeneity in structure, as well as the different growth conditions and pretreatments used.\textsuperscript{13−16} Due largely to the difficulty in the sample preparation, studying h-BN produced from scalable, integrated growth methods, such as CVD, has not been a priority for optical studies. However, the most promising method to produce h-BN with well-defined thickness over a large area is CVD.\textsuperscript{18,19} While layer thickness is known to strongly affect the band structure of h-BN,\textsuperscript{20} the optical properties of large crystalline domain monolayer h-BN are yet to be fully explored.

Here we focus on large areas (~100 mm\textsuperscript{2}) of continuous, single-crystalline (with domain sizes exceeding 100 μm\textsuperscript{2}) CVD-grown h-BN monolayers.\textsuperscript{18} These monolayers represent a homogeneous surface (without large variation in thickness, edges, or high density of grain boundaries) and thus present a well-defined model system to study defect emission in h-BN. The h-BN films are grown on a platinum catalyst and directly transferred using mechanical delamination onto glass substrates for the optical measurements. Our previous work has confirmed that crystalline domain size and continuity of the h-BN film are preserved following our transfer procedure.\textsuperscript{18,21,22}

It is typically difficult to find techniques that can locate and nondestructively probe such thin and optically transparent samples. Established structural techniques, such as high-resolution transmission electron microscopy (TEM)\textsuperscript{23} and scanning tunneling microscopy (STM),\textsuperscript{24} address small areas at a time and can induce new defects in the sample.\textsuperscript{25} Traditional optical techniques, such as confocal microscopy, typically do not enable uniform excitation fluence across a wide area, increasing the chance of non-linear effects.

To study this material, we use a custom-built spectrally resolved single-molecule fluorescence microscope, previously used in a biological context for multidimensional super-resolution imaging,\textsuperscript{26} that enables high-throughput measurements of relatively large areas (1250 μm\textsuperscript{2}) and simultaneous collection of spatial and spectral information. Surprisingly, we find that these h-BN monolayers produce emitters with high uniformity in emission wavelength. Unlike previous reports,\textsuperscript{27} we find one emission predominantly at 575 ± 15 nm. We also did not treat the h-BN with high-temperature argon annealing or electron irradiation to induce or stabilize emitters.\textsuperscript{13,28} We observe that the emitters display fluorescence intermittency (“blinking”) with “on” and “off” times described well by a power law distribution, similar to that previously shown in semiconductor nanoparticles or quantum dots.\textsuperscript{29−30} From an analysis of time-resolved spectral changes in conjunction with photodynamics, we find that single-emitter spectral diffusion is associated with blinking, leading us to conclude that a control of h-BN atmosphere or surface chemical treatment may improve spectral properties.

**RESULTS AND DISCUSSION**

**Power Law Blinking of Surface Emitters in h-BN.** The h-BN monolayer was grown following a protocol explained previously in detail.\textsuperscript{18} In Figure 1(b) the Raman spectra for

![Figure 1](image.png)
monolayer h-BN on SiO₂ is shown (red trace). A Raman peak at 1369 cm⁻¹ is seen and is not observed in the blank (black trace), indicating the presence of monolayer h-BN. In addition, TEM revealed the h-BN monolayers are made up of large (>100 μm²) aligned domains (Figure S1). We observe wrinkles in the h-BN on the platinum catalyst before transfer, in scanning electron microscopy (SEM) images (Figure S2), typical of h-BN monolayers. Combined, these results support that our preparation method created highly crystalline monolayer h-BN.

Wide-field super-resolution microscopy is used to illuminate large areas of the h-BN monolayers. Continuous-wave laser illumination is used to confine the excitation geometry (~200 nm axially) and excite the sample via an evanescent wave formed via total internal reflection (TIR) at the glass—air interface (Figure 1(a)). This geometry enables high sensitivity (signal/background ratio) typically used in single-molecule microscopy. The Stokes-shifted emitted light from the sample is collected via the same objective and imaged onto a (single-photon-sensitive) electron multiplied charge coupled device (EMCCD) camera. Images are stored as successive TIFF stacks with an exposure time of 100 ms (see Supporting Information for details).

Figure 1(c) shows a TIR fluorescence image (Z-stack of 1000 frames) of monolayer h-BN when excited with continuous-wave 532 nm laser light, in ambient air at room temperature; 532 nm (2.33 eV) is well below the band gap of h-BN (~6 eV) and therefore does not excite the bulk of the material, but does interact with the sub-band-gap defect states. Each diffraction-limited fluorescence puncta or point spread function (PSF) represents a single emitter. Each puncta can be analyzed in time (as shown in a time montage in Figure 1(d)) and can be fit to a 2D Gaussian to superlocalize its spatial position (typically 10–100 nm), where the localization precision is related to the fit error of the PSF and the number of photons (N) emitted:

$$\sigma_{\text{xy}} \approx \frac{\sigma_{\text{PSF}}}{\sqrt{N}}$$

For the emitters in h-BN, we obtain typical localization precisions ranging from 12 to 30 nm (Figure S6). We observe intermittency in the emission (Figure 1(d)), behavior that is typically attributed to a molecule or single quantum dot and representative of a single emitter, not an ensemble. As such, although the low photon count and instability of the emitters precludes us from recording second-order autocorrelation measurements, due to the discrete intensity trajectories observed, we contend that it is likely a large proportion of the blinking emitters observed are single emitters.

To characterize the fluorescence intermittency observed, in Figure 2(a) we plot the emission from a single emitter in monolayer h-BN over 100 s. The intensity of light from the emitter fluctuates over time. Using a threshold (details in Materials and Methods) to distinguish between “on” and “off” states, “on” and “off” times are calculated.
Figure 2(b) and (c) show the “on” and “off” time distributions obtained from histograms of “on” and “off” time, for 208 emitters measured across a total surface area of 0.018 mm² of three h-BN monolayer samples. Figure 2(c) and (d) represent over 13,000 data points each. The probability distribution for both times can be readily fitted to a power law of the form \( P(t) \approx t^{-m} \), where \( m_{\text{BN}} = -2.99 \) and \( m_{\text{OFF}} = -1.82 \). With a bin size of 100 ms, we find that the “off” times for the h-BN emitters are spread over 3 orders of magnitude in time (10⁻³–10⁴ ms) and the gradients to the power law fit are at the higher end for what has been observed for other nanoscale systems (1.7–2.2). Interestingly, such power law blinking behavior has been found and well studied in other nano-optical systems, such as quantum dots, GaN single-photon emitters, nanodiamonds, and single molecules, where it indicates either population of a “trap” or formation of a fluorescent inactive (“dark”) triplet state. We find here that both the “on” and “off” times show little dependence on laser intensity (inset in Figure 2(c)). No intensity dependence for the “off” times suggests the recovery mechanism of the emissive state is not light induced. A power law distribution of “off” times, as opposed to an exponential fit, points to a number of potential physical mechanisms for the formation of the nonemissive state, such as an exponential distribution of trap potential energy depths, fluctuating nonradiative decay rates, or a distribution of tunneling distances between photoinduced exciton and trap state. Characterization of the emitter photophysics may help us to understand the photoinduced dynamic processes occurring on the surface of h-BN in an ambient atmosphere.

Figure 3. (a) Schematic of the spectrally resolved measurement geometry. A collimated excitation source is totally internally reflected at the glass–air interface of the sample. Emitted light is directed through a tube lens and toward the diffraction grating and detector. (b) Z-stack of 1000 frames of the image recorded on the EMCCD. The spatial and spectral domains are contrast adjusted. (c) Zoomed-in view of the dashed box in the spectral region of image (b). The spectra are obtained from the first-order transmission through the diffraction grating. Spatial image of three emissive defects, obtained from the zeroth-order transmission through the diffraction grating. The integrated counts obtained for the circled emitter is 5.3 × 10⁴ counts/s. Each axis of the x,y scale bar equates to 1.5 μm. (e) Single emitter spectrum, middle spectrum in (c), for the middle emitter shown in (d).

Spectrally Resolved Wide-Field Imaging Reveals Monocolor Emitters. While blinking of single emitters is disadvantageous for quantum applications, it enables us to understand the photodynamics of individual surface-bound defects. blinking dynamics have been a critical tool in understanding trapping behavior, and designing methods of mitigating it, on the single-particle level for quantum dots. With a technique that has been deployed in research on quantum dots before, we perform spectrally resolved measurements to determine the nature of the emitters. Using a bespoke instrument we have previously reported, we simultaneously measure emitter position, blinking trajectories, and spectra. This is physically implemented on the setup described above, via the addition of a blazed diffraction grating, placed in the optical emission path before the image plane (Figure 3(a)). The two major diffraction orders (zeroth and first order) are projected onto different spatial regions of the EMCCD detector and recorded simultaneously (Figure 3(a) and (b)). The zeroth-order transmission corresponds to the spatial position of emitters on the surface of the h-BN (Figure 3(d)) and is used to superlocalize emitter position in x,y. The first-order diffracted light is the spectral information on the fluorescent signal (Figure 3(c)). The spectral resolution is related to the localization precision of the spatial image of the emitter (analogous to the slit width in a conventional spectrometer), thus related to the detected photons above background. We analyze all emitters with a fitted spatial resolution of <30 nm, corresponding to integrated photon counts of ~1.0 × 10³ photons/frame and a spectral precision in the range of 2–8 nm, with a typical value of 4 nm (Figure S5). The pixel-to-wavelength calibration of the detector is performed using fluorescent beads and takes account of spectral dispersion, as described in ref 26 and in Figure S4.
similarity in shape and ZPL position between the spectrum in Figure 3(e) and the reported spectra of single-photon-emitting defects in multilayer h-BN, we predict that the emission from the CVD-grown monolayer is likely to derive from defects of a similar structure. A control experiment of a blank glass slide subjected to the same polymer transfer confirmed that the emitters show a distinct emission profile from an organic contaminant (emission centered at 610−620 nm) (Figure S9) and that the organic contaminant has conclusively bleached within the first 50 s (Figure S8).

This inhomogeneous broadening is analogous to that observed in atomic or molecular transitions where radiating atoms or molecules interact differently with the environment, leading to a distribution of transition energies in the ensemble emission spectrum. The breadth of monolayer emission observed here (~190 meV), consistent with reports of the energy splitting observed between the ZPL and phonon sideband for h-BN defects, is likely to be the result of a broadened ZPL and phonon sideband, due to enhanced interaction with phonons. It has been shown that the full width at half-maximum (fwhm) of the ZPL in multilayer h-BN is exponentially dependent on temperature, consistent with in-plane phonon-assisted transitions. Interestingly, we find that despite the inhomogeneous broadening of single-defect linewidths, the ensemble population of emitters is more homogeneous than observed in thicker h-BN.

For every emission spectrum we resolve we fit the peak with a Gaussian and obtain the peak fluorescence emission wavelength. In Figure 4(a) and (b) we present histograms of the distribution of peak emission collected from h-BN monolayers from two independent growth runs, with insets showing the results for the two independent growths. In Figure 4(a) we present the histogram of all emission energies when emitters are sampled multiple times, termed "multiple sampling". The histograms reflect the total emission measured from the monolayers (>8000 emitter emission spectra). In contrast, for the histograms in Figure 4(b) the mean peak emission energy is reported for each emitter. Here emitters are sampled once, "single sampling". For all plots the bin size is 10 nm, slightly larger than the spectral resolution for the weakest emitters (8 nm). The data are not filtered to exclude any emission detected at 610−620 nm, a region we have identified as potentially due to contamination (Figure S9).

In both Figure 4(a) and (b) we observe a predominance of emission maxima at 575 ± 15 nm, a region repeatedly identified as a dominant ZPL for defects in multilayer and monolayer h-BN. Figure 4(a) and (b) give different information about the emitters’ behavior. The "multiple sampling" histogram reveals the ensemble-type emission, as would be recorded from a bulk photoluminescence measurement, while the "single sampling" shows the possible emission energies for the emitters. We can see that when each emitter is sampled once, the distribution of mean emission wavelength for emitters is weighted to higher energies (a grouping between 550 and 600 nm) than what we observe when we collect all emission maxima emitted from the monolayer. This suggests
that the most emissive emitters emit in a more narrow energy range, and at slightly lower energies, than the average emitter.

In Figure 4(c) we plot the distribution of average number of on-off-fluorescence intermittency cycles (“blinks”) for each emitter from one growth run, over 200 s of illumination. Each data entry here represents one emitter. As we can see, most emitters (87%) display less than 100 blinks. There is a small population (<2%) that display a very large number of blinks (>600). We assess the global photobleaching rate of the emitters and find that the average emitter total photon count has reduced by 60% after 200 s (Figure S8).

As our technique enables us to probe the behavior of individual emitters, we analyze the extremes of the population and can selectively interrogate both the very low and the very high blinking emitters, to assess the effect of blinking on spectral properties. In Figure 4(d) we plot all spectral maxima collected for emitters that show an average number of blinks greater than 600 and a median "off" time less than 30 s: the high blinking emitters. In Figure 4(e) we plot the same data for emitters that blink less than 100 times and have a median "off" time of less than 30 s: the low blinking emitters. This separation selects for emitters that are constantly blinking (~2% of total population) and those that exist in one state for long periods of time (~10% of total population). Indeed, we observe that emitters that show low blinking have a slightly narrower distribution of emission peaks (Figure 4(e)), while emitters that blink more show slightly more broadened emission (Figure 4(d)) and a strong dominance for emission between 570 and 580 nm. Combined, this analysis suggests that the most stable and emissive emitters show emission maxima in this narrow energy region.

Others have reported the presence of two distinct spectral classes of quantum emitters in h-BN: those that emit at ~570 nm and those that emit lower than 650 nm.13 Here we do not find evidence of a dominant low-energy emitter, which we attribute to the different growth and processing of the h-BN samples. We do not see any preference for emitters at edges or grain boundaries, and emitters appear to be randomly distributed across the monolayer, in a relatively low density (~0.04 emitters/μm²).48

The emitters on monolayer h-BN show a high degree of reproducibility when we compare individual growth runs; for growth 1, “multiple sampling”, 75% of emission spectra are at 575 ± 15 nm, for example, Figure 4(a) inset). We predict the difference in emission maxima between growth runs to be due to the varied degree of strain imparted into the monolayer during growth and/or transfer.16,51

These findings suggest the monolayers may be made up of predominantly one type of emitter that can interact with its environment to give a broadened line-width. To understand
more about this environmental interaction, we analyzed the spectrally resolved photodynamics. Across the h-BN samples, we observe that emitter behavior ranges from well-defined, telegraph-like on/off transitions (as displayed in Figure 2(a)) to emitters that transition between continuous distributions of emission intensities. To illustrate the range of behavior observed, in Figure 5(b) and (g) we show luminescence counts against time for two different emitters, emitter 1 and emitter 2. The fluorescence intensity trajectory for emitter 1 (Figure 5(b)) shows well-defined “on” and “off” levels in photon counts that clearly shows two clear intensity distributions (Figure 5(c)). The emitter is dark for the first 30 s before emitting, then subsequently enters one period of darkness before returning to the emissive state for the entirety of the measurement. Correspondingly, the color of emission is relatively constant (Figure 5(a)), with peak emission energy at 545 nm and fwhm of ~10 nm, displayed in the integrated (90–130 s) single-emission spectrum in Figure 5(c) and the distribution of all emission maxima in Figure 5(d).

In contrast, Figure 5(g) is the fluorescence trajectory for a second emitter that shows rapid blinking, a well-defined “off” photon count level, and a continuous distribution of photon emission events at higher photon counts. (Figure 5(k)). The emission maxima vary more than emitter 1 (Figure 5(h) and (i) show integrated emission spectra at different frames in the trajectory) and are on average lower in energy (centered at 560 nm) (Figure 5(j)). We find that the emitters across the samples display a range of behavior between these two examples (as demonstrated by Figure 4(c)), with no notable correlation with spatial position (previously observed in quantum dots).

Combined, these results show a correlation between blinking and spectral diffusion for single emitters in monolayer h-BN, closely reminiscent of the behavior observed in quantum dots, where perturbations in the local electric field on the surface of the quantum dots or systematic surface oxidation manifests as shifts in emission frequency. More generally, the demonstration of power law blinking and the overlap with quantum dot systems is significant; work over decades for quantum dot fluorescence intermittency to understand the complex photophysical phenomena at play and to exploit it to create rational devices. For example, the origin of blinking, and improving quantum yield have all been important in the development of quantum dots for different biological and energy applications. It may be possible to draw on this literature to enhance h-BN emitter properties. Certain approaches to control blinking, such as use of encapsulation or controlled atmospheres that prevent oxidation, may enable single-layer 2D materials to be viable candidates for future optical applications and should be explored.

CONCLUSION

We have shown that wide-field microscopies that are routinely used in biology can provide high-resolution, high-throughput, and multidimensional approaches to study the most challenging atomically thin material systems. In the search for deterministic, narrow-band, stable single photon emission, these results show that h-BN growth is important to consider. We show that CVD-grown monolayers with large crystalline domains represent homogeneous samples and produce emitters with highly reproducible emission maxima at 575 ± 15 nm. The emitter line shape is consistent with defects measured in thicker h-BN, and we propose they represent defects of the same structure. Our results show atomically thin h-BN can possess these emitters, without the need for high-temperature argon annealing or ion irradiation.

However, we show that emitters exposed to the surface suffer from disorder-inducing interactions, most likely with surface charges. Future work should involve control of surface species, via encapsulation for example. Methods of controlling surface contamination, without compromising optical extraction efficiency, will make atomically thin h-BN an attractive candidate for future optical quantum applications.

MATERIALS AND METHODS

Sample Preparation. h-BN was grown via a CVD method previously reported. In brief, h-BN is grown on a platinum catalyst by exposure at temperatures of 1000 °C to borazine. Continuous monolayers of h-BN form after a short period of growth. The samples are transferred onto the glass substrate using mechanical delamination. Hereby, a stamp is applied, and the h-BN layer can be directly peeled off the catalyst due to the weak interaction between the two. The stack consisting of stamp and h-BN is transferred onto a glass substrate and the stamp is removed. Prior to measurement all samples were annealed for 3 h at 500 °C in ambient atmosphere to remove all traces of organic contaminant from the transfer process.

Structural Techniques. Raman measurements were performed with a Renishaw inVia confocal Raman microscope. The TEM is carried out on a FEI Tecnai Osiris S/TEM under 80 keV. The diffraction mapping is done by taking a diffraction pattern at grid points with a step size around 1.8 μm in both the x and y directions. A selective aperture is used when taking the diffraction, so that the area of measuring has a ~1.4 μm radius. The x and y position is recorded by the reading from the piezo-stage. SEM images were taken on a ZEISS Gemini 300 VP scanning electron microscope using an acceleration voltage of 1 kV and an in-lens detector.

Optical Setup. Fluorescence imaging was performed using a home-built, bespoke inverted microscope (Olympus IX73) coupled to an EMCCD camera (Evolve II S2I, Photometrics, Tuscan, AZ, USA). The microscope was configured to operate in objective-type total internal reflection fluorescence (TIRF) mode. A 100 mW 532 nm continuous wave diode-pumped solid-state laser (LASOS Laser-technik GmbH, Germany) was directed off a dichroic mirror (Di02-R532-25×36, Semrock) through a high numerical aperture, oil-immersion objective lens (Plan Apochromat 60× NA 1.49, Olympus APON 60XOTIRF, Japan) to the sample coverslip. Total internal reflection was achieved by focusing the laser at the back focal plane of the objective, off axis, such that the emergent beam at the sample interface was near-collimated and incident at an angle greater than the critical angle for a glass/air interface for TIRF imaging. This generated a ~50 μm diameter excitation footprint with power densities in the range ~0.1–0.3 kW cm−2 at the coverslip, for all measurements. The emitted fluorescence was collected through the same objective and further filtered using a BLP01-532-25 long-pass filter (Semrock, USA) and a FF01-650/200-25 bandpass filter (Semrock, USA) before being expanded by a 2.5× relay lens (Olympus PE 2.5 × 125). Finally, a physical aperture and transmission grating (300 grooves/mm 8.6° blaze angle, GT13-03, Thorlabs) were mounted on the camera port path before the detector. The camera-to-grating distance was optimized using Tetraspeck beads (0.1 μm, T7297, Invitrogen) such that undiffracted and first-order diffraction was visible on the same image frame the fluorescence image was projected onto the EMCCD running in frame transfer mode at 20 Hz, with an electron multiplication gain of 250, operating at ~70 °C with a pixel size of 16 μm and automated using the open source microscopy platform Micromanager.

Images were acquired at a typical frame rate of 100 ms, and 1000–2000 frames were acquired. Each pixel on the image was equal to 107
nm. The coordinates corresponding to the localizations within the spatial part of the images were determined using the peak fit plugin for ImageJ.

Samples were measured in air, with the h-BN facing upward, illuminated from the other side of the glass.

**Spectral Calibration.** The location of the spectral domain on the EMMCCD chip is strongly dependent on the orientation and position of the diffraction grating, and so to ensure that the wavelengths determined were correct and therefore comparable, the instrument was calibrated before imaging each day and following realignment.

To calibrate the instrument, TetraSpeck beads were imaged by exciting at 405, 532, and 633 nm, and the emission was collected from 480 to 760 nm (Supplementary Figures 1–4) (100 frames were collected with a frame rate of 50 ms). The positions of the beads within the spatial domain of the image were determined using the PeakFit plugin (an ImageJ/Fiji plugin of the GDSC Single Molecule Light Microscopy (SMLM) package for ImageJ using a typical “signal strength” threshold of ~30. The fit result values are then saved as a text file in the directory containing the image for use in the subsequent spectral analysis below.

The spectral component of the image was analyzed using a custom-written Python code. The position of these three peaks allows both for the aberration correction (due to the relationship between these and the spatial positions of the beads) and for the pixel-to-wavelength ratio to be determined. For each localization (with coordinates $x$, $y$, frame number), the spectrum for each of the three peaks was averaged over a width of 3 pixels in the direction (to account for the spectra being wider than one pixel). Each intensity profile was fit to a Gaussian distribution to determine the center positions of the peaks. Fits that gave negative amplitudes, centers outside of the range, and widths of <1.5 pixels or >20 pixels were discarded.

The “on” and “off” times for all emitter trajectories were determined using the ImageJ PeakFit plugin. This plugin records localizations in a frame-by-frame manner if the localization meets a threshold signal (exact threshold parameters are given here). The output of the plugin is a text file with each localization detected recorded in each frame.

Emitters were deemed “on” if they were detected with the following PeakFit parameters: calibration: 107 nm/pixel, gain: 250, exposure time: 100 ms, initial stddev0: 2.0, initial stddev1: 2.0, initial angle: 0.0, smoothing: 0.5, smoothing 2 = 3, search width: 3, fit solver = [Least Squares Estimator (LSE)], fit_function = Circular, fit_criteria = [Least-squares error] significant_digits = 5, coord_delta = 0.0001, lambda = 10, max_iterations = 20, fail_limit = 3, include_neighbors, neighbor_height = 0.30, residuals_threshold = 0.50, shift_factor = 2, signal_strength = 50, width_factor = 2, precision = 30.

The code used to generate the “on” and “off” times counts the number of consecutive frames a localization with given $x$, $y$ coordinates is recorded (on time) and the number of consecutive frames a localization is not emissive between two emissive frames (off time).

A version of this article was previously uploaded to arXiv.39

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsnano.9b00274.

Detailed methods and experimental section as well as further characterization of the h-BN monolayers (PDF)

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