Grain Dependent Growth of Bright Quantum Emitters in Hexagonal Boron Nitride

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Abstract:

Point defects in hexagonal boron nitride have emerged as a promising quantum light source due to their bright and photostable room temperature emission. In this work, we study the incorporation of quantum emitters during chemical vapor deposition growth on a nickel substrate.

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Combining a range of characterization techniques, we demonstrate that the incorporation of quantum emitters is limited to (001) oriented nickel grains. Such emitters display improved emission properties in terms of brightness and stability. We further utilize these emitters and integrate them with a compact optical antenna enhancing light collection from the sources. The hybrid device yields average saturation count rates of $\sim 2.9 \times 10^6$ cps and an average photon purity of $g^{(2)}(0) \sim 0.1$. Our results advance the understanding of SPE incorporation during CVD growth and demonstrate a key step towards compact devices for achieving maximum collection efficiency.

1. Introduction:

Atomic defects in solid state hosts that act as single photon emitters (SPEs) are promising hardware components for a range of emerging quantum technologies,^[1] such as quantum network nodes, quantum repeaters, and quantum key distribution. Despite rapid advances culminating in these proof of principle demonstrations, reliable fabrication techniques and subsequent integration with nanophotonic components remains challenging.^[2]

Recently, layered van der Waals materials such as hexagonal boron nitride (hBN) have been explored as hosts of SPEs.^[3] This class of materials is of a particular interest as their layered nature provides avenues for nanoscale manipulation and reliable integration with photonic devices.^[4] hBN SPEs are especially promising due to the stability of the host,^[5] room temperature operation,^[6] outstanding optical properties,^[7] and the presence of optically detected magnetic resonance signals.^[8]

SPEs in hBN can be engineered through chemical vapor deposition (CVD),^[9] molecular beam epitaxy,^[8a, 10] or metal organic vapor phase epitaxy.^[8a] Bottom up fabrication encompasses two primary advantages over top down fabrication methods. The first is the ability to fabricate cm scale thin films with controlled thickness that are ideal for transfer and incorporation with nanophotonic platforms.^[11] The second is the ability to manipulate the photophysical properties of the incorporated SPEs during growth, such as defect density,^[12] emission energy,^[12] and the fabrication of homogeneous ensembles.^[8a]

In this work, we report on low-pressure CVD growth of hBN on polycrystalline Ni foils. We demonstrate that SPEs are selectively incorporated only in hBN grown on (001) Ni, despite confirming that growth occurs on all grain orientations. Remarkably, the SPEs incorporated on (001) Ni display superior emission intensity and stability compared to previous CVD growth studies on metallic catalysts.^[9, 13] Consequently, we transfer the thin-film hBN to a planar dielectric antenna to enhance its collection efficiency. Our hybrid device yields average saturated count rates of $\sim 3 \times 10^6$ counts/s and a high emission purity $g^{(2)}(0) \sim 0.1$. Our results advance the understanding of SPE formation in layered materials and provide a unique and promising route to deterministic SPEs for quantum photonics.

2. Results and Discussion

Figure 1a displays a schematic illustration of the CVD hBN growth on polycrystalline Ni foil. The growth of hBN via low-pressure CVD was performed following an established protocol.^[9a] Figure 1b displays a Fourier-transform infrared spectrum recorded from as-grown multilayer hBN films. The spectrum is from a large area that contains all Ni grain orientations, and displays the three

characteristic FTIR active stretching modes,^[14] confirming the growth of thin-film hBN. Figure 1c displays an optical image of a selected region of the Ni film covered with thin-film hBN. A clear contrast in the colors of the different Ni grains can be observed, consistent with previous reports.^[15] Figure 1d displays the same region analyzed by scanning electron microscopy, where the grain boundaries are similarly visible.

To determine the orientation of each Ni grain, we analyzed the region with electron beam secondary diffraction (EBSD), Figure 1e. The image displays the orientation of the Ni domains at the surface, and it can be seen that (111), (101), and (001) oriented grains are distributed randomly across the foil with individual grains showing surfaces areas of $\sim 10\text{-}50\mu\text{m}^2$. To probe for local variations in the material quality of the as-grown hBN film (i.e. within a particular single crystal grain), we collected the Raman spectra from (111), (101), and (001) oriented grains displayed in Figure 1f. hBN displays only one prominent Raman active stretching mode – an E_{2g} mode at $\sim 1367\text{cm}^{-1}$.^[16] Note that the collection parameters were held constant across the measurements, and the individual peaks were normalized to the (001) spectrum, permitting a direct comparison of relative intensity. For (101) and (001) grains we observe a strong resonance at 1368.1cm^{-1} and 1367.5cm^{-1} , respectively. The signal is $\sim 20\%$ weaker on the (101) grain, while both display a near-identical peak full width at half maximum (FWHM) of $\sim 33.5\text{cm}^{-1}$. The similar widths suggest similar material quality,^[17] while the hBN film may be slightly thicker on the (001) grains.^[16] In contrast, we only observe a weak Raman signal for hBN grown on (111) oriented Ni grains. The presence of the E_{2g} mode centered at 1367.4cm^{-1} confirms that hBN is present on (111) grains. The signal is ~ 5 times weaker, suggesting that less material has been grown. However, the peak displays a narrower FWHM (18.7cm^{-1}) than observed on the (101) and (001) grains, indicating a slightly higher material

quality of the hBN grown on (111) grains. Additionally, we transferred the hBN thin-film to a Si/SiO₂ substrate observing a continuous film results from the growth. Atomic force microscopy (AFM) was used to characterize the thickness of the transferred film, finding thickness variations from ~6-12 nm, see Supporting Information. Continuous film coverage and variations in the film thickness are consistent with the conclusions drawn from the Raman data, as well as with the inhomogeneity of hBN grown on polycrystalline Ni observed previously in the literature.^[18] We further discuss the implications of the grain dependent growth rate, and underlying mechanism below.

We next characterize the photoluminescence (PL) of hBN grown on different Ni grains to examine potential variations in SPE incorporation. Note that all measurements were performed with the hBN adhered to the Ni growth substrate. Figure 2a displays a selected region where the grains are sufficiently small to be investigated in-tandem, within the field of view of our optical setup. Utilizing a custom-built scanning confocal PL setup, we mapped a selected area (indicated by the black dashed line in figure 2a). Figure 2b displays the resulting confocal scan, where we observe a clear dependence of emission intensity from the hBN film on the orientation of the underlying Ni substrate. Specifically, hBN on (001) grains displays a bright fluorescence, whilst that on (101) and (111) grains is drastically reduced.

Figure 1. CVD growth of hBN on polycrystalline Ni foil. **a.** Schematic illustration of hBN growth on Ni (111), (101), and (001) grains, where orange spheres represent SPEs which are only observed on (001) oriented Ni grains. **b.** FTIR spectrum from the as-grown hBN on nickel, showing the LO and TO active modes. Optical **(c)** SEM **(d)** and EBSD **(e)** images of as-grown hBN on Ni. Inset in e displays the legend relating the grain orientation to color. **f.** Raman spectroscopy of hBN on different Ni grains (111), (101), (001). The spot of each collection is marked with a white circle in panel e.

We further checked for the presence of SPEs in each subsequent grain orientation, by recording the spectrum from localized excitation spots. Figure 2c shows a representative spectrum of an SPE located in a (001) Ni grain, displaying the characteristic line shape of hBN SPEs in the visible spectral range. The emitter has a zero-phonon-line (ZPL) at ~573nm and a phonon side-band (PSB) consisting of 2 peaks at ~644nm and ~653nm.^[19] To probe the nature of the emission we performed

second order auto-correlation measurements on the emission peak, and obtained $g^{(2)}(0) < 0.5$, confirming the quantum nature of the emission, inset Figure 2c. While SPEs are easily located within corresponding (001) Ni grains, we were unable to locate any narrow emission peaks resembling hBN SPEs in either (101) or (111) Ni grains within the entire 125x125 μm scan area in Figure 2b.

Figure 2. Grain dependent incorporation of hBN SPEs. **a.** EBSD map demonstrating the Ni grain orientation in the characterized area. The black (white) dashed box corresponds to the area mapped in **b** (**d**) respectively, while the white circle marks the SPE in **c**. **b.** Confocal PL mapping (532nm 300 μW) of the black dashed area. The white circle marks the SPE displayed in **c**. **c.** Spectrum of

representative SPE localized at the white circle in a and b. Inset displays the $g^{(2)}(\tau)$ from the same emitter. **d.** Wide-Field imaging of white dashed area marked in a. A high density of SPEs on (001) Ni grains is apparent while no SPEs appear in alternative grain orientations.

To directly visualize the grain dependent incorporation of SPEs and to evaluate the density of SPEs in (001) grains we additionally performed wide-field imaging, as shown in figure 2d. The imaged area corresponds to the white dashed area in figure 2a. Each localized emission center is attributed to an SPE. These results confirm the high density of SPEs incorporated in (001) Ni grains, while SPE are not observed on alternative grain orientations. Additional areas characterized *via* wide-field imaging all demonstrate that incorporation of SPEs is limited to (001) oriented grains, see Supporting Information.

One potential explanation for these observations is grain dependent variations in the surface structure or roughness of differently oriented Ni grains. In order to probe this, we performed both AFM mapping of the hBN on Ni as well as SEM imaging to probe for nanoscale structural differences between the grain orientations, see Supporting Information. SEM imaging reveals clear differences in the nanoscale structure across the grain types, specifically that (111) and (101) grains display a ridged wave like surface pattern, while (001) grains display a tiled appearance, with the respective patterns occurring within a single crystal grain. By correlating these images with wide-field PL maps taken from the same areas we can rule out the nanostructure of the Ni surface as being primarily responsible for our observations. Similarly, by correlating surface roughness measured with AFM to grains showing SPE emission we can rule out surface roughness as the origin of our observations, as

the roughest grain measured was a predominantly (001) oriented grain showing effective SPE incorporation, see Supporting Information.

Having precluded surface roughness and nanoscale surface texture as the origin of selective incorporation of SPEs on Ni (001) grains, we reflect on the current understanding of the growth mechanism of hBN on Ni. It has been well documented that interactions between the metallic catalyst and the precursor are of critical importance to the growth process,^[17, 20] while interactions between the substrate and the as-grown hBN film can also influence the properties of the first hBN layer.^[21] It has previously been postulated that variations in the surface energy,^[15] or the sticking coefficient of the precursor compounds,^[18b] on different Ni grain orientations is responsible for the observed growth rate dependencies. However, more recent work has confirmed that the role of diffusion and segregation of precursor species within the Ni catalyst also play a central role.^[18a] Boron in particular is known to readily diffuse through bulk Ni at high temperatures, effecting hBN growth,^[22] while both Ni self-diffusion and impurity diffusion (B, N, etc.) are tied to vacancy generation and mobility within the nickel bulk catalyst.^[23] In fact, at temperatures near that used during growth in this study (1030°C), the diffusivity of atoms both within the bulk and as near surface impurities in nickel is highest on the (001) plane.^[24]

It is also pertinent to consider the role of impurity atom diffusion, such as carbon and oxygen. In fact, interstitial carbon atoms in Ni have been shown to alter the kinetics and hBN growth process during CVD,^[25] and carbon has also been utilized to aid in the decomposition of ammonia borane on the nickel surface.^[26] Importantly, this decomposition process is thought to proceed through a boron oxide intermediate which prevents sub-surface diffusion into the Ni by B and N species.^[26] More generally surface oxidation is also known to be of critical importance for the CVD

growth of 2D materials.^[27] To probe experimentally the degree of surface oxidation for individual Ni grains we performed an annealing process of the Ni foil, identical to that used prior to growth, but instead of performing hBN growth we instead analyzed the pre-growth surface oxidation levels on individual Ni grains by correlating EBSD mapping with elemental mapping via energy dispersive X-Ray spectroscopy (EDS). We observed that the concentration of oxygen on (001) grains is significantly reduced compared to those on (101) or (011) grains, suggesting that diffusion will be enhanced on (001) Ni grains, see Supporting Information. Furthermore, previous studies have demonstrated that modifying catalyst diffusion effects can control both the density and emission energies of hBN SPEs during CVD growth on copper.^[12]

As a result, we propose that grain-dependent variations in atomic diffusion processes provide the most straightforward interpretation of the selective incorporation of SPEs on Ni (001) grains. The consequences of the proposed mechanism are twofold. The first is through modified feeding rates of B and N to the surface, a parameter known to influence the formation energies of atomic defects.^[28] The second is *via* the incorporation dynamics of heteroatom impurities such as carbon, which has recently been linked to SPEs in hBN.^[8a, 29] Our interpretation is strengthened by noting that while grain dependent diffusion effects are also observed in CVD of hBN on Cu, they are relatively minor in comparison to Ni showing negligible variations in the observed growth kinetics.^[30] This is consistent with previous reports demonstrating that SPEs are incorporated homogeneously across various Cu grain types,^[9a] suggesting grain based diffusion variations are not significant enough to affect SPE incorporation on Cu.

Having established the grain dependent incorporation of SPEs on Ni (001), we explore in further depth their optical properties. Figure 3a displays a histogram of ZPL energies for 84 separate

SPEs localized on Ni (001). Emitter ZPLs are found to be clustered around 580 nm, with ~87% <600nm, in agreement with previous studies on SPEs in CVD grown hBN materials.^[9a, 9b] The inset for Figure 3a shows a typical stability trace for a Ni (001) emitter at an excitation power of 300 μ W, demonstrating a stable emission rate despite the nanoscale dimensions of the film (~10 nm). This stability is notably in contrast to previous studies on CVD hBN growth on a range of metal foils, including copper, which displayed a high affinity for blinking and bleaching.^[9, 13]

Figure 3. Optical properties of hBN SPEs incorporated on Ni (001) grains. a. Histogram plotting the ZPL positions of 84 analyzed SPEs on Ni (001) grains, with a 5nm bin width. The emission lines are clustered around 580nm. The inset shows a representative stability trace from an SPE at 300 μ W **b.** The lower panel is a scatter plot comparing the relative brightness of 10 representative SPEs on Ni (001) grains (blue triangles) and on Cu (red triangles). Dashed lines indicate the average count rates for each set of 10 SPEs. The upper panel records the brightest SPEs observed from Ni (001) grains, which are separated due to their higher relative brightness.

Next, we compare the overall intensity of the emitters grown on Ni and Cu foils. Figure 3b (bottom panel) plots the emission intensities of 10 representative SPEs characterized on Ni (001) grains (blue) contrasted with 10 SPEs grown on Cu foil (red). To confirm that differences in the collection efficiency from Cu and Ni were not substantial, we performed laser reflection experiments from multiple spots on both annealed Cu and Ni, and well as from each metal with hBN grown on top, finding no significant effect on observed reflection efficiency, see Supporting Information. Each sample was grown under otherwise identical growth conditions, and their brightness compared by evaluating the fitted spectrometer counts under equivalent excitation conditions (300 μ W, 10sec acquisition). Brightness was evaluated *via* spectral comparison (as opposed to photon counting) to reduce the potential contribution of varying background emission rates between the samples. In order to ensure spectrometer collection efficiency did not influence the findings, only SPEs with a ZPL at 580 ± 10 nm were analyzed. SPEs incorporated on Ni (001) display an average ~ 2.5 -fold enhancement in emission compared to those on copper (dashed lines Figure 3b). The enhancement fluorescence of SPEs on Ni (001) planes is likely the result of favorable environmental influences—such as a decreased prevalence of nearby charge traps or additional point defects—as quantum efficiency and brightness are known to vary even locally,^[31] however, the exact mechanism responsible for emission rate fluctuations in hBN SPEs remains unknown. Figure 3b (top panel) displays the three brightest emitters characterized on Ni (001) grains, the brightest of which displayed a fluorescence signal roughly 10x the average SPE observed on Ni (001) with an estimated count rate of $\sim 7.6 \times 10^6$ cps, see Supporting Information. The extreme brightness of this particular SPE highlights the potential for ultra-bright emission centers fabricated *via* bottom up growth.

The improved emission rate and stability of SPEs incorporated on Ni (001) grains encompassed within a nanometer scale host material (<10 nm) makes this system appealing for integration into planar dielectric antennas (PDAs) which can enhance collection efficiency from the source^[32] providing the source is encapsulated within an extremely thin host material. PDA structures have been successfully implemented in the past for efficient light extraction from single molecules in organic crystals,^[33] quantum dots in thin polymer films,^[34] and Nitrogen vacancy centers in diamond.^[35] These devices are inherently broadband,^[36] robust against fabrication imperfections, and compatible with a variety of single photon sources.

Figure 4a depicts a schematic illustration of the PDA design used in this work, consisting of a glass coverslip capped with a 250 nm layer of MgF₂ on top of which the hBN is transferred (cf. methods). To ensure optimal operation of the hybrid structure, we must consider the refractive index (n), the thickness of the hBN layer, as well as the emission energy (λ_{SPE}) of the SPE. Specifically, for a hBN layer thickness ($n_{\text{hBN}}=1.8$)^[37] approaching $\lambda_{\text{SPE}} / (2 * n_{\text{hBN}})$, the appearance of propagating modes within hBN will occur. In this case the hBN layer will act as a 2D waveguide created by the MgF₂/hBN/air stack ($n_{\text{MgF}_2} < n_{\text{hBN}}$, $n_{\text{air}} < n_{\text{hBN}}$), and the light coupling to these unwanted modes cannot be channeled into the collection optics, effectively lowering collection efficiency. Therefore, for optimal operation of the antenna, the hBN film thickness must be $\ll \lambda_{\text{SPE}} / (2 * n_{\text{hBN}})$. In practice, hBN layer thicknesses ≤ 30 nm are optimal to negate all unwanted channeling effects, making epitaxial hBN films, in this case ~ 10 nm thick, ideally suited for PDA integration. Figure 4b shows the simulated collection efficiency and relative power density from an in-plane linearly polarized dipole in hBN integrated with the PDA, calculated in accordance with previously published methods.^[38] We find that more than 84% of the fluorescence emission can be channeled within a collection angle of

70°, which can be easily collected by using an oil immersion objective (NA of 1.46). The collection efficiency from Ni was also simulated for comparison, demonstrating that the PDA represents a calculated enhancement of ~2 in the power that reaches the far field for collection.

To quantify performance of the hBN/PDA device, we characterized the saturated count rate and purity of 3 representative SPEs. Figure 4c displays the saturation curves of 3 such SPEs. To provide an upper bound for the saturated emission rate from each SPE, we measured the count rate on the SPE and subtracted the background emission recorded from ~1μm away. Each recorded saturation curve was fit with the equation $f(P) = I_{\infty} \frac{P}{P_{Sat} + P}$, where I_{∞} is the saturated count rate, and P_{Sat} is the saturation power. The obtained fits for SPE 1, 2, and 3 yield saturated count rates $I_{\infty} = 3.92 \pm 0.23 \times 10^6$ cps, $2.27 \pm 0.05 \times 10^6$ cps, and $2.59 \pm 0.19 \times 10^6$ cps. The corresponding saturation powers are $P_{Sat} = 39.8 \pm 4.8 \mu\text{W}$, $25.2 \pm 1.4 \mu\text{W}$, and $36.0 \pm 5.6 \mu\text{W}$ respectively. The high-count rates, displaying average values $I_{\infty} \sim 2.9 \times 10^6$ cps at a very low saturation power of ~34 μW, demonstrate the promise of the integrated hBN/PDA device structure. We note, that the count rates observed on the hybrid hBN/PDA device represent an average quantity, which is much higher than observed on Ni, with the exception of the rare extremely bright centers.

Figure 4. Integration of hBN films with a planar dielectric antenna. **a.** Schematic representation of the hBN/PDA structure. **b.** Simulated power density (light red/gray region) and collection efficiency (red/gray line) as a function of the collection angle for a hBN SPE excited on the antenna (red) and on nickel (gray). For the PDA the maximum collection efficiency is $\sim 84\%$ at collection angles of $\sim 70^\circ$, while on nickel the maximum collection efficiency is $\sim 42\%$ at collection angle of $\sim 90^\circ$. The inset displays the simulated emission profile for an hBN SPE on both surfaces with a dipole oriented in plane. **c.** Saturation curves for 3 separate hBN SPEs excited with a 532nm laser with powers from 0-66 μW . **d.** Corresponding $g^{(2)}(\tau)$ collections for the 3 emitters in figure 3c.

Figure 4d displays the associated $g^{(2)}(\tau)$ for SPE 1, 2, and 3, each collected with an excitation power of 10 μ W. We note in all cases, no additional spectral filtering or background subtraction was performed to provide a quantitative measurement of the emission purity. Fitting the collections for SPE 1, 2, and 3 yield $g^{(2)}(0)=0.08$, 0.18, and 0.07 respectively, confirming the high purity of the emission, see Supporting Information. The high-count rates, stability, and emission purity observed from the hybrid hBN/PDA device, provide a simple, yet promising and scalable platform for emerging quantum technologies.

3. Conclusions

In summary, we have demonstrated that hBN SPEs grown on polycrystalline Ni foil are incorporated preferentially in hBN on Ni (001) grains. We propose that the origin of the effect is variations in the diffusion and subsequent supply of atomic species during growth, a parameter known to vary with the grain orientation of the Ni catalyst. Intriguingly, controlling these diffusion effects on a nanometer scale could enable spatially controlled SPE incorporation during growth. These emitters display an enhanced fluorescence intensity ($\sim 2.5\times$) compared to those incorporated during hBN growth on Cu, with select SPEs on Ni (001) displaying an emission intensity 10x greater than average. Finally, leveraging the nanoscale dimensions of the hBN thin films (~ 10 nm), we incorporate them in a planar dielectric antenna to maximize collection efficiency. Characterization of SPEs in the hybrid device yield average count rates of $\sim 2.9 \times 10^6$ cps at a saturation power of $\sim 34 \mu$ W, and a high emission purity of $g^{(2)}(0) \sim 0.1$. The hybrid devices are promising for room-temperature

integrated quantum photonics with hBN and pose an efficient strategy to maximize collection efficiency from other two-dimensional quantum light sources.

4. Methods

LPCVD growth of hBN: Low-pressure chemical vapor deposition (LPCVD) was used to grow thin-film hBN on nickel and copper foils (Sigma-Aldrich), following a previously established method.^[9a] In brief, Brasso (a commercial metal polisher) was used to polishing the foil and remove oxide residuals from the surface. The foil was then cleaned by sonication in dilute HCl, acetone, and isopropyl alcohol for 15 mins each followed by rapid drying under an N₂ stream. The cleaned foil was instantly placed in a quartz tube at the center of a ceramic tube furnace. Ammonia borane powder (Sigma-Aldrich) was loaded into a metal crucible and attached to the system. Prior to growth the sample was annealed in an Ar/H₂ environment for 2 hours to increase the grain size of the foil. Growth was performed for 45 minutes at a furnace temperature of 1030°C, a pressure of 2 Torr, a 50 sccm flow rate of Ar/H₂, a precursor temperature of 95°C.

Planar dielectric antenna fabrication: The antenna was fabricated by deposition of a 250 nm layer of MgF₂ on top of a glass coverslip of thickness 170 $\mu\text{m} \pm 5 \mu\text{m}$ (Marienfeld-Superior), using a Univex400 Thermal Evaporator and a deposition rate of 0.4 nm/s at 25° C and a vacuum of $\leq 10^{-7}$ mbar. Note, the MgF₂ layer thickness is not critical and it can be off by several tens of nanometers without affecting the performance of the antenna.

Assembly of hBN/PDA Device: The hBN film was then transferred to the antenna using a poly(vinyl)alcohol (PVA) assisted transfer method.^[39] Briefly, the hBN on Ni sample was coated with a

PVA/glycerol solution, and was used to coat the entire hBN/Ni surface. The sample was then dried on a hot plate at $\sim 40^{\circ}\text{C}$ for 2 hours, before peeling off the hBN/polymer layer and deterministically placing the stack on top of the PDA antenna. The sample was then heated at 120°C for 30 minutes to increase adhesion between the hBN and the MgF_2 , before placing in warm milli-Q H_2O overnight to remove the polymer layer. The next day the assembled hBN/PDA device was then placed in a UV-Ozone cleaner to remove any remaining polymer residuals before PL analysis.

Raman spectroscopy: Raman spectroscopy was performed on an In-Via confocal Raman (Renishaw) system with a 633 nm excitation source. Calibration of the Spectrometer was carried out using a Si substrate to 520 cm^{-1} . The peaks were then fitted to a Lorentzian line profile.

Fourier transform infrared spectroscopy (FTIR): Infrared spectra were measured using an attenuated total reflectance (ATR) setup (Nicolet 6700, Thermo-Scientific).

Electron beam secondary diffraction (EBSD): EBSD mapping and scanning electron microscope imaging was performed on a Thermo Fischer Scientific Helios G4 feg SEM. The EBSD measurements were recorded using a 5 keV electron beam at a 10 mm working distance with a 70° sample tilt. The signal was collected using an Oxford C-Nano Electron backscatter diffraction detector and processed using AZtec.

Confocal PL on Ni: PL studies were carried out using a custom-built scanning confocal microscopy with continuous wave (CW) 532 nm laser (Gem 532, Laser Quantum Ltd.) as excitation. The laser was directed through a 532 nm line filter and a half-waveplate and focused onto the sample using a high numerical aperture (100 $\times$, NA = 0.9, Nikon) objective lens. Scanning was performed using an X-Y piezo fast steering mirror (FSM-300). The collected light was filtered using a 532 nm dichroic mirror

(532 nm laser BrightLine, Semrock) and an additional long pass 568 nm filter (Semrock). The signal was then coupled into a graded-index multimode fiber (0.22 NA), where the fiber aperture of 62.5 μm serves as a confocal pinhole. A flipping mirror was used to direct the emission to a spectrometer (Acton Spectra Pro, Princeton Instrument Inc.) or to two avalanche photodiodes (Excelitas Technologies) in a Hanbury-Brown and Twiss configuration, for collection of spectra and photon counting, respectively. Correlation measurements were carried out using a time-correlated single photon counting module (PicoHarp 300, PicoQuant). All of the second-order autocorrelation $g^{(2)}(\tau)$ measurements were analyzed and fitted without background correction unless otherwise specified.

Wide-field-imaging: For Wide-field imaging experiments, a lens system was used to focus the excitation beam at the Fourier plane of the objective, creating a collimated illumination spot to collectively excite a large area of the sample. 532 nm CW excitation at a power of 5 mW was performed in conjunction with a 0.55 NA objective, allowing for an approximate field-of-view of $\sim 50 \times 50 \mu\text{m}^2$. Quantum emitters in various hBN samples were recorded on an EMCCD (Andor iXon Ultra 888) with an exposure time of 200 ms for 100 acquisitions.

PL analysis of hBN/PDA device: The hBN/PDA device was investigated using a confocal excitation scheme. Excitation was performed *via* a CW laser COHERENT-Sapphire 532-200 through a 1.46 NA oil immersion objective (Zeiss alpha-Plan-Apochromat 100x) in an inverted microscope setup. A dichromatic mirror is used to separate the excitation beam from the fluorescence signal. The emitted fluorescence is then sent through a telescope in combination with a movable 75 μm pinhole for further spatial filtering and reduction of the background. The fluorescence beam is then guided into a Hanbury-Brown and Twist (HBT) configuration where the stream of single photons is split by a 50/50 Pellicle beam splitter and then send into two high quantum efficiency APDs

(Lasercomponents). The photon events are then recorded using a HydraHarp 400-module from PicoQuant and analyzed with a software (Symphotime 64) also from PicoQuant. Alternatively, a silver-protected mirror (M1) can be flipped to redirect the fluorescence signal into a single emitter sensitive camera Hamamatsu ORCA-Flash4.0 for alignment and visual inspection of the sample. Another mirror (M2) can be flipped to monitor the fluorescence spectrum of the sample. The sample is mounted on a 3-axis piezo actuator Tritor 400 Piezosysteme Jena for precise alignment of the emitters with respect to the laser focus. For exploring large sections of the sample, the 3D piezo stage is mounted on a 2D manual-micro-positioner. Figure S1 displays a simplified schematic of the setup.

Supporting Information:

Supporting Information is available from the Wiley Online Library or from the author.

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59 word TOC summary:

This work explores the growth of hexagonal boron nitride (hBN) on polycrystalline nickel foil, finding that quantum emitters are incorporated only on nickel grains oriented in the (001) direction. These emitters display extremely bright and photostable quantum emission, a first for few-layer hBN films. Finally, these quantum sources are integrated with a compact optical antenna to maximize collection efficiency.

TOC Graphic (55 x 50 mm):

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