Identifying carbon as the source of visible single-photon emission from hexagonal boron nitride

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Single-photon emitters (SPEs) in hexagonal boron nitride (hBN) have garnered increasing attention over the last few years due to their superior optical properties. However, despite the vast range of experimental results and theoretical calculations, the defect structure responsible for the observed emission has remained elusive. Here, by controlling the incorporation of impurities into hBN via various bottom-up synthesis methods and directly through ion implantation, we provide direct evidence that the visible SPEs are carbon related. Room-temperature optically detected magnetic resonance is demonstrated on ensembles of these defects. We perform ion-implantation experiments and confirm that only carbon implantation creates SPEs in the visible spectral range. Computational analysis of the simplest 12 carbon-containing defect species suggest the negatively charged V5C0 defect as a viable candidate and predict that out-of-plane deformations make the defect environmentally sensitive. Our results resolve a long-standing debate about the origin of single emitters at the visible range in hBN and will be key to the deterministic engineering of these defects for quantum photonic devices.

single defects in solids have become some of the most promising front-runner hardware constituents of applications in quantum information technologies and integrated quantum photonics. Substantial effort has been devoted to isolate and deterministically engineer such defects in wide-band-gap materials such as diamond and silicon carbide. This collective effort resulted in spectacular proof-of-principle demonstrations ranging from quantum networks to spin–photon interfaces, while simultaneously and steadily leading to understanding the fundamental-level structures of these defects.

Recently, hexagonal boron nitride (hBN) has emerged as a promising host material for defects which display ultrabright single-photon emission (SPEs). They exhibit remarkable properties: a strong response to applied strain and electric fields (Stark shifts), stability under high pressure and elevated temperatures, potential for resonant excitation above cryogenic temperatures and addressability via spin-selective optical transitions. Yet, despite the numerous experimental characterizations and in-depth theoretical attempts to model their possible crystalline structure, the nature of these defects remains unknown.

Part of the challenge stems from standard hBN bulk crystal synthesis via high pressure and high temperature not being amenable to the deterministic control of impurity incorporation. This is aggravated by the induced impurities often segregating and forming regions of inhomogeneous defect concentration. In addition, the two-dimensional, layered nature of hBN makes ion implantation difficult to control. These limitations have precluded identifying the exact origin of the single-photon emission in the material.

Here we address this problem by carrying out a detailed study surveying various hBN samples grown in different laboratories by metal–organic vapour-phase epitaxy (MOVPE) and molecular beam epitaxy (MBE). We find compelling evidence that to observe photoluminescence from SPEs the inclusion of carbon atoms in hBN is required. By systematically growing samples with different carbon concentrations, we show that the carbon content determines whether the photoluminescence signal originates from an ensemble of emitters (high carbon concentration) or isolated defects (low carbon concentration). Defect ensembles are demonstrated to display room-temperature optically detected magnetic resonance (ODMR). We carry out multispecies ion-implantation experiments on both MOVPE films and exfoliated hBN, showing that only carbon implantation creates SPEs and that the density of emitters scales directly with the implantation dose of carbon. Our results are supported by rigorous modelling analysis of carbon-related defects.

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The rationale was to understand whether the single defects are intrinsic or extrinsic defect. However, it is clearly a spin-carrying defect complex. The energy detuning between the ZPL of the ensemble and phonon sideband (PSB) peak is ~176 meV on average (Extended Data Fig. 1). X-ray photoelectron spectroscopy (XPS) was used to quantify the incorporation of carbon (Extended Data Fig. 2). Figure 1bc demonstrates a near-linear correlation between C–B (C–N) bonding and increasing TEB flux, with C–B bonding being roughly an order of magnitude more prevalent than C–N bonding. Preferential formation of C–B bonds follows logically from noting the boron species are introduced with three pre-existing bonds to carbon. The PL intensity of the resulting ensemble emission likewise displays a linear correlation with carbon concentration (Extended Data Fig. 3). Based on these results, we advance that the SPE emission at ~580 nm in hBN probably originates from a carbon-related defect complex.

Figure 1d shows the ODMR spectra recorded from the TEB 60 ensemble. The highly symmetric shape of the signal does not reveal a structure that would allow a clear assignment to a specific intrinsic or extrinsic defect. However, it is clearly a spin-carrying defect, likely with a spin state higher than 1/2. By varying the static magnetic field, we measure resonances at ~523, ~668.5 and ~815.4 MHz for B = 19, 24 and 29 mT, respectively. A value for g, of

## Table 1 | Epitaxial hBN samples with varying carbon concentrations

| Sample | Abbreviation | Growth method and details | SPE PL | Additional information |
|--------|--------------|---------------------------|--------|------------------------|
| MOVPE hBN (TEB flux 10) | MOVPE hBN (TEB 10) | MOVPE on sapphire, precursors TEB and ammonia, TEB flow 10 µmol min⁻¹, H₂ carrier gas, 1,350 °C | Isolated SPEs, ZPLs predominantly ~585 ± 10 nm | -40 nm thick |
| MOVPE hBN (TEB flux 20) | MOVPE hBN (TEB 20) | MOVPE on sapphire, precursors TEB and ammonia, TEB flow 20 µmol min⁻¹, H₂ carrier gas, 1,350 °C | Dense and uniform ensemble of SPEs with ZPL ~585 nm, PSB ~630 nm | -40 nm thick |
| MOVPE hBN (TEB flux 30) | MOVPE hBN (TEB 30) | MOVPE on sapphire, precursors TEB and ammonia, TEB flow 30 µmol min⁻¹, H₂ carrier gas, 1,350 °C | Dense and uniform ensemble of SPEs with ZPL ~585 nm, PSB ~630 nm | -40 nm thick |
| MOVPE hBN (TEB flux 60) | MOVPE hBN (TEB 60) | MOVPE on sapphire, precursors TEB and ammonia, TEB flow 60 µmol min⁻¹, H₂ carrier gas, 1,350 °C | Dense and uniform ensemble of SPEs with ZPL ~585 nm, PSB ~630 nm | -40 nm thick |
| MBE hBN on sapphire | Undoped MBE hBN on sapphire | MBE on sapphire, boron flux from e-beam source (300 W), boron in BN crucible, N₂ flow 2 sccm, growth temperature 1,250 °C. | No SPEs present | -20 nm thick |
| MBE hBN on sapphire with carbon crucible | Carbon-doped MBE hBN on sapphire | MBE on sapphire, boron flux from e-beam source (210 W), boron in carbon crucible, N₂ flow 2 sccm, growth temperature 1,250 °C. | Semi-isolated SPEs, ZPLs 570–770 nm, density ~5–8 µm⁻² | -18 nm thick |
| MBE hBN on SiC (0° Si face) | Undoped MBE hBN on SiC (0°) | SiC (Si face, orientation-on) MBE on SiC, boron flux from HT Knudsen source at 1,875 °C, N₂ flow 2 sccm, growth temperature 1,390 °C | Very few SPEs, density ~1 SPE in 40 µm² | -3 nm thick |
| MBE hBN on SiC (8° Si face) | Undoped MBE hBN on SiC (8°) | SiC (Si face, orientation 8°-off) MBE on SiC, boron flux from HT Knudsen source at 1,875 °C, N₂ flow 2 sccm, growth temperature 1,390 °C. | Isolated SPEs, ZPLs 575–735 nm, density ~3–5 µm⁻² | -7 nm thick |
| HOPG → hBN conversion | Converted hBN | HOPG is placed in a radiofrequency induction furnace at 2,000 °C, N₂ gas is mixed with thermalized B₂O₃ powder facilitating conversion | Dense and uniform ensemble of SPEs with ZPL ~580 nm, PSB ~630 nm | Bulk |

See Methods for additional synthetic details.
The quantum nature of the emission was confirmed by measuring the second-order autocorrelation function; the value of $g^{(2)}(r=0) < 0.5$ (Fig. 1d inset) is conventionally attributed to a single photon source with sub-Poissonian emission statistics. We measured the ZPL wavelength of 77 SPEs in the MOVPE hBN (TEB 60) sample, finding that ~78% of the emitters are located at 585±10 nm and 95% at wavelengths <600 nm (Extended Data Fig. 6), consistent with previous studies on epitaxially grown hBN. The typical line shape of these emitters at room temperature is also consistent with previous studies, including the ZPL and a PSB centred at ~1.77 meV from the ZPL energy. This suggests that when the carbon concentration is sufficiently low, individual quantum emitters can be isolated. Their optical properties and spectral distribution are consistent with those observed in samples with higher carbon doping, with the difference merely being due to the density of emitters.

To further confirm that carbon-based defects are responsible for SPE emission from hBN we analysed a series of hBN samples grown by a different method: MBE. Fig. 2a displays the PL spectrum observed from undoped MBE hBN grown on sapphire substrate. The resulting PL signal was relatively low; no SPEs could be found despite the material being of good quality as shown by a clear hBN Raman line. However, when the elemental boron source was placed inside a carbon crucible—with otherwise identical growth conditions—we observed the appearance of sharp spectral lines, shown in Fig. 2b. The carbon crucible used for e-beam evaporation of the boron shows clear signs of sidewall etching, which suggests that carbon was present in the gas phase during growth.

The carbon-doped MBE growth resulted in a high density of emitters such that we could not isolate at single sites. We instead probed the polarization dependence of particular emission peaks by placing a polarizer in the collection path. Figure 2c shows one such collection, where emission from a ZPL at ~577 nm is linearly polarized, with the PL intensity dropping to the background level when the polarizer is perpendicular to the polarization direction of the probed emitter.

We next explored MBE growth of hBN on silicon carbide (SiC), investigating different crystal orientations: specifically, with the top Si face-on (0°) and slightly off (8°). Representative spectra from
both sample types (Si at 0° and at 8°) are displayed in Fig. 2d. When growth was performed with the Si face at 0°, only a single SPE peak was located across a 40 µm² scan. In contrast, when the Si face is oriented at 8° we again find a high density of SPE incorporation. The incorporated SPEs display a similar ZPL distribution to the orientated at 8° we again find a high density of SPE incorporation.

We attribute the incorporation of these SPEs during hBN growth during growth. The samples were then annealed in high vacuum (1,000 °C, 6 torr, 2 h), and the same set of measurements was performed. The remaining ~20% of SPEs within the implanted region display similar line shapes and phonon coupling to those for the samples synthesized while increasing carbon content during growth.

Figure 3a shows the confocal scan of the TEB 10 sample after carbon implantation, but prior to annealing, where a TEM grid with 50 µm² square apertures was used as a mask. The implanted region is labelled I, while the masked region is labelled II. Figure 3b displays spectra collected from emitters within the implanted region (I), and a representative g²(τ = 0) < 0.5, confirming the quantum nature of the emission from these centres. Figure 3c displays a representative emitter from the masked region (II), showing the typical line shape of the ZPL and the PSB peaks found in TEB 10 films, with the corresponding g²(τ = 0) shown to the right.

Inside the carbon-implanted region, most emitters (~80%) display narrow ZPL peaks (~5 nm full-width at half-maximum (FWHM)) and extremely weak PSBs compared with the typical ZPL/PSB found in these TEB 10 films (Extended Data Fig. 7). The remaining ~20% of SPEs within the implanted region display similar line shapes and phonon coupling to those for the emitter in Fig. 3c and are attributed to pre-existing SPEs in the region. Our results indicate that the sharp emission lines belong to SPEs created via implantation of carbon ions. The reasons for the observed narrow line shape and the minimal phonon coupling are explored further via computational modelling below.

The samples were then annealed in high vacuum (1,000 °C, <10⁻⁴ torr, 2 h), and the same set of measurements was performed. As shown in Fig. 3d, the implanted regions are still visible; however, they show variations in PL intensity. This effect is likely due to ion scattering around the mask edges and vacancy diffusion—which have been observed for implantation in diamond. The PL spectra from three different areas are shown in Fig. 3e–g, and correspond to (I) the implanted region of high PL intensity, (II) the implanted region of lower PL intensity and (III) the masked region of the film.

Figure 3e displays a representative spectrum from inside region I, where we found broad emission similar to those observed in the high TEB flux growths. This emission is confirmed to be due to an ensemble of SPEs as the corresponding g²(τ) measurements
show no anti-bunching despite the associated ZPL/PSB structure. A similar spectral signature is observed consistently throughout region I, again implying the creation of an ensemble of carbon-based SPEs. Figure 3f displays a representative spectrum from the implanted region II, where we again observe luminescence with a similar line shape. The overall ensemble signal
remains homogeneous in this region, although appears less dense and bright, and the $g^{(2)}(r)$ measurement shows a value of ~0.75, confirming the presence of fewer emitters within a confocal spot. Note that in both implanted areas (I and II) we no longer observe the narrow emission lines with low phonon coupling found before annealing. Finally, Fig. 3g displays a representative spectrum from region III (masked area), showing a typical ZPL and PSB profile with a $g^{(2)}(0)<0.5$. Control experiments implanting silicon and oxygen with otherwise identical conditions were also performed, but the emitters, either singles or ensembles, were not observed (Extended Data Fig. 8).

To further study SPE formation via ion implantation we performed dose-dependent experiments with carbon fluences over the range $1 \times 10^{11} - 10^{14}$ ions cm$^{-2}$, while oxygen and silicon implantation at $1 \times 10^{13}$ ions cm$^{-2}$ served as a control. Both MOVPE (TEB 10) hBN and exfoliated pristine hBN flakes (HQ Graphene) were used. The samples were analysed via wide-field imaging, allowing for the direct visualization of the resulting SPE density.

Figure 4a shows representative images from the exfoliated hBN flakes before and after annealing. The results demonstrate unambiguously that emitter creation scales with the dose of carbon implantation in both cases, which directly confirms the creation of SPEs. Only a few emitters are formed pre-annealing, even at higher doses, but a direct correlation between SPE formation and implantation fluence is clearly evident post-annealing.

Figure 4b shows a direct comparison for carbon, oxygen and silicon ion implantations at a dose of $1 \times 10^{13}$ ions cm$^{-2}$. For the MOVPE samples implanted with oxygen and silicon we observe a similar SPE density to pristine TEB 10, while carbon implantation considerably increases the density. In the exfoliated samples, only carbon implantation results in the direct formation of single emitters at a high density.
The main defect candidates were considered: CB, C N, V N CB and V BCN. CAM-B3LYP density functional (see Supplementary Information and converted from raw emission $\lambda$ region I (blue, Fig. 3b C-implanted) and region II (red, Fig. 3c masked). The observed spectra are shown after correction for instrument response functions.

The only feasible emission source is the (1)4$_B$ $\rightarrow$ (1)4$_A$ transition in V$_8$C$_N$, with fully optimized three-ring, one-layer and three-ring, three-layer structures shown in e, along with the allowed in-plane-perpendicular electric polarization vector. f, predicted band shape (black dashed line, basic CAM-B3LYP three-ring, one-layer model; black solid line, QM/MM EOM-CCSD ten-ring, three-layer out-of-plane distorted model) compared with observed ones from region I (blue, Fig. 3b C-implanted) and region II (red, Fig. 3c masked). The observed spectra are shown after correction for instrument response functions and converted from raw emission E($\lambda$) to band shape $\lambda$E($\lambda$) plotted versus energy $h\nu = hc/\lambda$, displaying broadening to a resolution of 0.01eV. The predicted spectra are both too low in energy and too broad compared with the experimental ones, but the assignment is within computational uncertainty.

Given the large number of possible defect candidates considered, we proceeded by eliminating unsuitable ones by benchmarking our calculations to known experimental properties. We focused on three well-established experimental features of the SPEs, a ZPL energy transition of $\approx$2.1eV$^{32,33}$, a fast excited state lifetime of $\approx 2$–6ns$^{34}$ and a high quantum efficiency$^{35}$. Accordingly, computational results were filtered to reproduce, first, a CAM-B3LYP-calculated lowest-energy transition of 1.6–2.6eV (based on the expected worst-case computational error, calibrated for this method to be $\pm 0.5$eV$^{36}$), and second, an oscillator strength exceeding 0.1, compatible with the observed short photoluminescence lifetime and high quantum yield. Few defects have lowest transition energies in this range, and most transitions are predicted to have oscillator strengths 100th of this or much less. Based on these considerations, only two candidates remain of interest amongst the options considered: the (1)4$_B$ $\rightarrow$ (1)4$_A$ transition in V$_8$C$_N$, and the (2)5$_B$ $\rightarrow$ (1)4$_B$, transition in V$_8$C$_N$. Of these, V$_8$C$_N$ is immediately eliminated as its spectral band shape and most other properties are highly inconsistent with observed features; hence we focus on V$_8$C$_N$.

The ground state of V$_8$C$_N$ is predicted to be (1)4$_A$, with unpaired electrons in the a$_1$(s), b$_2$(s) and b$_3$(s) orbitals. Four low-energy excited states are predicted, of which the lowest-energy one would need to be (1)5$_B$. One-layer models predict that this state undergoes out-of-plane distortion which lowers the energy. The distortion, however, can be either removed or enhanced once multilayer models are considered. This transition has dominant $a_1$(s) $\rightarrow$ $b_2$(s) character, polarized in-plane and perpendicular to the defect’s C$_N$ axis (see Fig. 5e), with an oscillator strength exceeding 0.1. Figure 5f compares calculated emission bandshapes $E(\nu)$/$\nu^2$ (obtained as the raw emission scaled by wavelength to the fifth power) with experimentally observed spectra from Fig. 3. The (1)4$_B$ $\rightarrow$ (1)4$_A$ emission is predicted to be slightly lower in energy and much broader. The calculated width is environment dependent (Fig. 5f) and dominated by how the calculations perceive torsional changes at the defect associated with light emission that generate low-frequency phonons. The observed spectra in region I are indicative of such effects, but their magnitude is reduced to one-third. The observed spectra in region II are very different, primarily manifesting the effects of activation of BN-stretch phonons instead.

Electronic structure calculations

To gain further insight into the structure of the carbon defect, we searched for defect transitions from which the observed photoemission could originate. To do so, time-dependent density-functional theory$^{39}$ (TD-DFT) calculations were performed using the CAM-B3LYP$^{40}$ density functional (see Supplementary Information for extensive details). These are supported by calculations using the HSE06 density functional$^{41}$ and the advanced equation-of-motion coupled cluster singles and doubles (EOM-CCSD)$^{42}$ methods. Four main defect candidates were considered: C$_N$, C$_N$$_2$ V$_8$C$_N$ and V$_8$C$_N$ (Fig. 5a–d) in their neutral, negative (−1) and positive (+1) charged states. Two spin manifolds were considered for each (either singlet and triplet or else doublet and quartet), as well as at least ten excited states of each type. Calculations were performed using three-ring, five-ring and ten-ring model compounds containing one or three hBN layers, to account for the effects of the host matrix on the defect. Figure 5e displays the three-ring, one-layer and three-layer model for V$_8$C$_N$. Calculations on ten-ring systems were performed using a mixed quantum-mechanics/molecular-mechanics (QM/MM) scheme, utilizing an AMBER$^{43}$ potential fitted to mimic CAM-B3LYP results on five-ring hBN.

Figure 4c shows two spectra recorded from the localized emission spots in the carbon-implanted exfoliated samples, before and after annealing. Green and red circles in Fig. 4a mark the position of the recorded spectra in each case. Additional wide-field imaging and spectral characterization for exfoliated and MOVPE hBN are displayed in Extended Data Figs. 9 and 10.

In light of the implantation results, we briefly consider a potential ancillary role of carbon. This could occur through the stabilization or charge state modification of alternative defects, as well as modification of the material Fermi level. Critically, our implantation results allow us to rule out these possibilities. The creation of SPEs prior to annealing occurs only for carbon implantation (that is, not with silicon and oxygen implantation), despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation, despite clear evidence of increased vacancy creation excluding the secondary role of carbon as that of silicon and oxygen implantation.
The most important shortcoming of the proposal of this defect as the dominant hBN SPE would be that intense absorption is predicted in only one polarization, whereas experiments suggest that higher-energy absorptions exist with alternate polarization\(^\text{4,14}\). However, of the 24 defect manifolds considered herein, it is the only one to remain of interest. More complex carbon-cluster defects, including, for instance, C\(_2\)CN and C\(_2\)CV, have been considered as alternatives\(^\text{14,15}\). In summary, we have presented rigorous experimental results to confirm the central role of carbon in hBN quantum emitters in the visible spectral range. We compared samples grown by MOVPE, MBE and HOPG conversion. All methods exhibited a direct correlation between the introduction of carbon as a precursor/substance and the formation of SPEs. Furthermore, MOVPE growth enabled us to deterministically control carbon incorporation and vary the density of the quantum emitters from single to ensembles and observe room-temperature ODMR. We have also generated SPEs using direct ion implantation of carbon and showed that their density scales with the implantation dose. Employing a TD-DFT method, we proposed the negatively charged V\(_{\beta}\)CN as a suitable transition to explain the observed results. Our results will accelerate the deployment of visible quantum emitters in hBN into quantum photonic devices and will advance potential strategies for the controlled engineering of quantum emitters in van der Waals crystals.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41563-020-00850-y.

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Methods

MOVPE. hBN layers were grown on commercially available 2 inch sapphire substrates using MOVPE, as described in ref. 27. TEB and ammonia were used as the boron and nitrogen precursors, respectively, while hydrogen was the carrier gas. The precursors were introduced into the reactor as short alternating pulses, in order to minimize parasitic reactions between TEB and ammonia. hBN growth was carried out at a reduced pressure of 85 mbar and the growth temperature was set to 1,350 °C. In the present study, the TEB flux was varied from 10 to 60 μmol min⁻¹ to study the effect on carbon incorporation on sub-band-gap luminescence from the hBN films. For ion implantation, PL and SPE measurements, centimetre-sized hBN films were transferred from sapphire on to SiO₂/Si substrates, using water-assisted self-delamination3. The thickness of the hBN films was also measured using atomic force microscopy (AFM), as described in the Supplementary Information. XPS was used to determine the impurity levels in the as-grown MOVPE hBN films, with an on–off modulation. The resonant condition was changed with the external magnetic field by mounting a permanent magnet below the sample. With an on–off modulation, the microwaves were driven with an off–on modulation. The resonant condition was changed with the external magnetic field by mounting a permanent magnet below the sample. Additional information is available from the corresponding author upon request due to their size. Source data are provided with this paper.

Data availability

Source data for most experimental and theoretical data for this work are provided. Confocal maps and wide-field images are available from the corresponding author upon request due to their size. Source data are provided with this paper.

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Author contributions

N.M. and I.A designed the experiments. N.M., J.R.R., C.B. and I.A. wrote the manuscript with contributions from all co-authors. N.M. performed experimental measurements and data analysis. J.R.R. and M.J.F performed the computational calculations. D.C., C.J. and H.H.T performed ion implantation and MOVPE growth. T.S.C., C.M., P.H.B and S.V.N. performed MBE growth. H.L. and A.Z fabricated the HOPG to hBN conversion samples. A.G. and V.D. performed ODMR experiments. I.A., C.B. and M.T. supervised the project. All authors discussed the results and contributed to the manuscript.

Competing interests

The authors declare no competing interests.

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Extended Data Fig. 1 | MOVPE hBN ensemble ZPL/PSB detuning. **a**, An ensemble spectrum taken from MOVPE hBN (TEB 30) showing the energy separation between the ensemble of ZPLs to the resolved phonon sidebands. The detuning from the ZPL centroid at 2.122 eV to the LO₁ phonon mode at 1.961 eV is 161 meV. While the detuning from the ZPL to the LO₂ mode at 1.927 eV is 195 meV. **b**, An ensemble spectrum taken from a different confocal spot of the MOVPE hBN (TEB 30) sample where both the first and second order PSBs can be observed. The ZPL ensemble centroid is positioned at 2.116 eV, and the first PSB (which appears as a single peak due to the convolution of the LO₁ and LO₂ modes) is centered at 1.937 eV, a detuning of 179 meV. Additionally, a dimmer broad peak can be observed at lower energy spanning from roughly 1.789 eV to 1.725 eV, which is due to the second order phonon modes which are comprised of three independent emissions, 2LO₁, LO₁+LO₂, and 2LO₂.
Extended Data Fig. 2 | XPS C1s spectra from MOVPE hBN TEB series. MOVPE hBN samples with increasing TEB flow. 

- **a**, MOVPE hBN (TEB 10).
- **b**, MOVPE hBN (TEB 20).
- **c**, MOVPE hBN (TEB 30).
- **d**, MOVPE hBN (TEB 60).
Extended Data Fig. 3 | PL intensity of MOVPE hBN with increasing TEB flow vs the B-C + B-N Bonding %. The integrated intensity of the ZPL peak from each MOVPE sample (as plotted in Fig. 1a) is plotted against the bonding percentage of C-B + C-N as determined by XPS. For TEB 10,20,30 we see an almost perfectly linear trend. For TEB 60 we observe a slightly reduced intensity increase, likely the result of non-radiative decay pathways induced by an increasingly defective material.
Extended Data Fig. 4 | Extraction of g value from room temperature ODMR (TEB 60). ODMR resonance frequencies as a function of applied magnetic field, extracted from Fig. 1e. The data points are fit with equation S1 and yield an extracted g value of ~ 2.09.
Extended Data Fig. 5 | Temperature dependent ODMR of MOVPE (TEB 60) hBN. We recorded the ODMR contrast from the highly carbon doped MOVPE (TEB 60) sample at four temperatures between 295-13 K. A similar FWHM of the resonance suggests the broadening is dominated by unresolved hyperfine interactions.
Extended Data Fig. 6 | Histograms of ZPL positions from various epitaxial sources. 

**a**, 77 SPEs characterized in MOVPE hBN (TEB 10) display ZPLs clustered around 585 ± 10 nm.

**b**, 248 SPEs characterized from CVD hBN on copper display ZPLs clustered around 580 ± 10 nm, reproduced with permission from ref. 28.

**c**, 65 SPEs characterized in carbon doped MBE hBN on sapphire display ZPLs ranging across the visible spectrum.

**d**, 26 SPEs characterized in undoped MBE hBN on silicon carbide displaying ZPLs ranging across the visible spectrum.
Extended Data Fig. 7 | ZPL FWHM comparison of SPEs located inside the C implanted region to as-grown single photon emitters in MOVPE hBN (TEB 10). Blue triangles correspond to SPEs analyzed from as-grown MOVPE hBN (TEB 10). Red triangles correspond to SPEs located within the C implanted region of the same sample. The implantation created SPEs show a nearly 4 fold reduction in average linewidth.
Extended Data Fig. 8 | Spectra of MOVPE hBN (TEB 10) samples implanted with oxygen and silicon. Implantations were done at a dose of $10^{13}$ cm$^{-2}$ and an energy of 10 keV, using a TEM grid with 50 µm$^2$ apertures as a mask. 

**a**, Typical spectra observed in the oxygen-implanted region pre-annealing, showing only background emission the $V_\gamma$ peak ~ 800 nm are observed.

**b**, Characteristic spectrum from oxygen implanted region post annealing, where the only spectral signature observed is a broad peak at ~ 630 nm.

**c**, Typical spectra observed in the silicon-implanted region pre-annealing, showing only background emission the $V_\gamma$ peak ~ 800 nm are observed.

**d**, Characteristic spectrum from silicon implanted region post annealing, with the only spectral signature observed is a broad peak at ~ 630 nm.
Extended Data Fig. 9 | Wide-field imaging and spectral analysis of exfoliated hBN implanted with carbon prior to annealing. The scale bar in each is 2 µm. **a**, Un-implanted exfoliated hBN reference sample. **b**, Exfoliated hBN implanted with carbon at a fluence of 1×10^{11} ions/cm^2. **c**, Exfoliated hBN implanted with carbon at a fluence of 1×10^{12} ions/cm^2. **d**, Exfoliated hBN implanted with carbon at a fluence of 1×10^{13} ions/cm^2. **e**, Exfoliated hBN implanted with carbon at a fluence of 1×10^{14} ions/cm^2. **f**, Un-implanted MOVPE hBN reference sample. **g**, MOVPE hBN implanted with carbon at a fluence of 1×10^{11} ions/cm^2. **h**, MOVPE hBN implanted with carbon at a fluence of 1×10^{12} ions/cm^2. **i**, MOVPE hBN implanted with carbon at a fluence of 1×10^{13} ions/cm^2. **j**, MOVPE hBN implanted with carbon at a fluence of 1×10^{14} ions/cm^2.
Extended Data Fig. 10 | Wide-field imaging and spectral analysis of MOVPE hBN implanted with carbon. The scale bar in each is 2 µm. a, Un-implanted MOVPE hBN reference sample. b, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{11}$ ions/cm$^2$. c, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{12}$ ions/cm$^2$. d, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{13}$ ions/cm$^2$. e, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{14}$ ions/cm$^2$. f-j samples were annealed at 1000 °C for 2 hours under vacuum ($<1 \times 10^6$ mbar). f, Un-implanted MOVPE hBN reference sample. g, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{11}$ ions/cm$^2$. h, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{12}$ ions/cm$^2$. i, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{13}$ ions/cm$^2$. j, MOVPE hBN implanted with carbon at a fluence of $1 \times 10^{14}$ ions/cm$^2$. 

| Carbon Implanted MOVPE hBN (Pre Annealing) |  
| No Implant | $10^{11}$ | $10^{12}$ | $10^{13}$ | $10^{14}$ |
|---|---|---|---|---|
| a | b | c | d | e |

| Carbon Implanted MOVPE hBN (Annealed) |  
| No Implant | $10^{11}$ | $10^{12}$ | $10^{13}$ | $10^{14}$ |
|---|---|---|---|---|
| f | g | h | i | j |

Normalized Intensity

Wavelength (nm)

Normalized Intensity

Wavelength (nm)