Single-spin resonance in a van der Waals embedded paramagnetic defect

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A plethora of single-photon emitters have been identified in the atomic layers of two-dimensional van der Waals materials1–8. Here, we report on a set of isolated optical emitters embedded in hexagonal boron nitride that exhibit optically detected magnetic resonance. The defect spins show an isotropic $g$-factor of -2 and zero-field splitting below 10 MHz. The photokinetics of one type of defect is compatible with a hyperfine coupling of ~10 MHz. Its narrow linewidth and inhomogeneously broadened magnetic resonance spectrum differs significantly from the known spectra of in-plane defects. We determined a hyperfine coupling of ~10 MHz. Its angular dependence indicates an unpaired, out-of-plane delocalized spin, consistent with a paramagnetic defect. We found that the photodetected magnetic resonance (ODMR) of these defects recorded at 5 K and 300 K exhibit magnetic field-dependent PL. A magnetic field was applied externally with a permanent magnet. The magnetic field strength $B$ as well as its direction and magnitude varied during the experiment. The ODMR spectra were recorded by applying microwaves (MWs) through a 20-μm-thick wire placed in close proximity to the emitter (Fig. 1h). A magnetic field was applied externally with a permanent magnet. The magnetic field strength $B$ as well as its direction were determined by performing ODMR on single negatively charged defects in hexagonal boron nitride.

Our research was performed on single-crystal h-BN supported on a SiO$_2$ substrate. The details of the sample fabrication are given in the Methods section. Three different defects were identified that exhibit a measurable ODMR signal and they will be referred to as defect D1, D2 and D3 (Extended Data Figs. 1 and 2). Figure 1a displays the distinct photoluminescence spectra of these defects recorded at 5 K. D1 possesses a relatively narrow optical linewidth, and also shows clear phonon replica (Extended Data Fig. 3) associated with the phonon spectrum of the h-BN crystal. The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5). The spectrum of defect D1 is identical to that of a defect reported previously. The latter exhibited magnetic field-dependent PL at room temperature. We also show in Fig. 1c that the background-corrected autocorrelation values at zero delay $g^2(0)$ for D1 and D2 are 0.59 and 0.22, respectively (Extended Data Fig. 2 and Supplementary Section 5).

ODMR spectra were recorded by applying microwaves (MWs) through a 20-μm-thick wire placed in close proximity to the emitter (Fig. 1h). A magnetic field was applied externally with a permanent magnet. The magnetic field strength $B$ as well as its direction were determined by performing ODMR on single negatively charged defects in hexagonal boron nitride.

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charged nitrogen vacancy (NV\(^{-}\)) centres in a diamond placed in the vicinity of the h-BN flake (Extended Data Fig. 5). Figure 1b–d shows the ODMR spectra recorded at 5 K for defects D1, D2 and D3, respectively. A single ODMR peak is observed for each defect shows the ODMR spectra recorded at 5 K for defects D1, D2 and D3, respectively. The peak frequencies correspond to magnetic fields of approximately 154, 290 and 200 G, respectively. The solid lines are Gaussian fits to the data with a g-factor close to 2 (see below). The ODMR linewidth shows some variation among the defects, and ranges between 25 and 35 MHz. Two important distinctions between these ODMR data and those reported previously for spin-bearing defects, such as three-boron-centre defects\(^{12}\), are the narrowness and single-peak nature of the resonance. We note, however, that our findings are consistent with the recently observed ODMR of defect ensembles\(^{16–18}\) generated by carbon irradiation.

To address the electronic level structure and photophysical properties of these defects that lead to the observation of a magnetic resonance, we analysed the kinetics of the optical excitation and emission processes to derive the most relevant relaxation parameters and determine whether the spin-bearing states are metastable or rather in the electronic ground state of the system. Specifically, the response of the defects to different MW and optical excitation sequences was studied, and is exemplarily shown for defect D2 in Fig. 2.

The steady-state ODMR contrast, that is, the change in fluorescence upon spin resonance, is plotted as a function of the incident laser power in Fig. 2b. It initially increases, peaks at about 20% for a laser power of 10\(\mu\)W, and then starts to drop when the laser intensity is ramped up further. Such behaviour of the ODMR contrast hints at a ground-state origin of the ODMR and some spin polarization of the ground state rather than the involvement of metastable states (see Extended Data Fig. 6). In the latter case, one would expect a monotonic increase in the ODMR effect in accordance with the observed monotonic increase in the fluorescence intensity as a function of the optical excitation power (Supplementary Section 10).

More information on the electronic level structure of the defect was gathered by carrying out the three different sequences of optical and MW excitation summarized in Fig. 2c and referred to as excitation sequences S1–S3. For sequence S1, the sample was only excited optically with a 3-ms laser pulse. The readout pulse occurred after a variable delay time \(\tau\). For sequence S2, MW radiation was incident during the entire delay time \(\tau\). Because the MW excitation was absent from sequence S1, only the population decay of the optically excited states and specifically the metastable states to the ground state was measured. As is apparent from Fig. 2d, the fluorescence, and thus ground-state population (fluorescence recovery), increases as the delay time \(\tau\) of the readout pulse increases (Fig. 2d). Hence, during the first optical pulse, part of the population is shelved into a relatively long-lifetime metastable state. The extracted decay constant of 100\(\mu\)s is consistent with the decay component derived from \(g^2\) measurements (Supplementary Section 10). We therefore interpret this time constant as population decay from an excited metastable state. The variation in signal with delay time obtained for sequence S2 is also plotted in Fig. 2d. The recovery curve is essentially identical to the previous case, indicating that the ODMR does not occur in the metastable state and that some polarization is present in the ground state instead, in line with our interpretation of the data in Fig. 2b.
addition to metastable states m₁ and m₂. Such long-lived states have forced us to postulate the existence of at least one very long-lived state in while retaining the decay rate from the metastable states. This forces we observed that the fluorescence intensity reduces significantly were applied simultaneously during the optical initialization pulse, level diagram is, however, incomplete. When both MWs and laser optimal conditions before a repump mechanism becomes more also reflected in the simulation of the laser power dependence of .

Supplementary Section 10), it is possible to estimate a spin–lattice relaxation process.

All of the above observations are compatible with the level diagram model presented in Fig. 2a. Under laser excitation and in the absence of MWs, an initial mixed population in the spin singlet ground states g₁ and g₂ is pumped to the excited states e₁ and e₂. On relaxation, the latter preferentially populate the g₁ state through non-radiative decay via metastable states m₁ and m₂. When the MWs are switched on, the populations in g₁ and g₂ are modified. This is also reflected in the simulation of the laser power dependence of the ODMR contrast plotted as a solid line in Fig. 2b. It peaks under optimal conditions before a repump mechanism becomes more dominant at higher laser power (Supplementary Section 10). The level diagram is, however, incomplete. When both MWs and laser were applied simultaneously during the optical initialization pulse, we observed that the fluorescence intensity reduces significantly while retaining the decay rate from the metastable states. This forces us to postulate the existence of at least one very long-lived state in addition to metastable states m₁ and m₂. Such long-lived states have been reported to play a role in h-BN single defects (for a detailed discussion, see Supplementary Section 10).

We also performed ODMR in pulsed mode (sequence S3), repeated 20K times for each measurement point to ensure a quasi-steady state despite the presence of a long-lived state. The delay time τ between the initial laser pulse and the MW pulse was kept greater than the excited-state lifetime (∼1 ns). Yet, the sequence yields a significant ODMR contrast, as seen in Fig. 2c. This eliminates the possibility of ODMR originating directly from the excited states. Moreover, as the time delay increases, the contrast decreases, and from the observed T_{deph} = 3 µs (τ, in Fig. 2c, also see Supplementary Section 10), it is possible to estimate a spin–lattice relaxation time T₁ value of ∼10 µs. This is in agreement with the estimated order of magnitude, discussed later (see Fig. 4g).

We next turned our attention to the magnetic field dependence and linewidth analysis of the ODMR. With the help of an NV-centre in the diamond in the measurement set-up (Extended Data Fig. 5), it is possible to determine both the strength and the angle of the external applied magnetic field. Figure 3b shows the ODMR spectrum of defect D2 for a magnetic field of 288 G. The ODMR peak is inhomogeneously broadened (Extended Data Fig. 7) as revealed by comparison with various model functions. Figure 3c illustrates the shift in the ODMR peak to higher frequency on increasing the in-plane magnetic field B₀. The central frequency and width of the resonance are described by a spin Hamiltonian H comprising the interaction of a single electron with N nuclear spins:

H = gₑB₀S + SDS + ∑_{i=1}^{N} (gᵢB₀Iᵢ + SAIᵢ + IᵢQIᵢ).

Here, gₑ and gᵢ are the electron and nuclear g-factors, S and I are the electron and nuclear spin operators, respectively, D is the zero-field splitting (ZFS), A is the hyperfine tensor and Q is the nuclear quadrupole interaction and βₑ,n here represents the electron and nuclear magnetic moments. We first focused on the purely electronic components of the Hamiltonian and analysed the observed line shift. From a least-squares linear fit to the magnetic field-dependent resonance frequency in Fig. 3c,d, we extracted the slope or gₑ-factor of 2.06, close to the value for the free electron.
Monitoring the ODMR signal at different angles allowed us to determine its anisotropy (Supplementary Section 7). The g-factor follows the relation \( g_{\perp} + \Delta g \cos^2 \theta \), where \( \Delta g = g_{\parallel} - g_{\perp} \) is the difference between the in- and out-of-plane g-factor, and \( \theta \) is the angle with respect to the plane. Within our experimental uncertainty, \( \Delta g \) (the g-factor) is isotropic. Limited data on the anisotropy of the g-factor of h-BN defects are available in the literature. Moore and Singer, for example, reported \( \Delta g = 9.5 \times 10^{-4} \), which is below our detection accuracy. Our value of \( g \) is of similar magnitude to those measured in the EPR analysis of h-BN. g-factors larger than 2 (2.0024 to 2.0032) are reported for carbon-rich materials compared to boron due to a larger spin–orbit coupling\(^{12,14}\). The intercept at zero field of the linear fit to the magnetic field dependence of the resonance frequency in Fig. 3c yields the ZFS parameter \( D \). For D1, the scatter in the data does not allow a meaningful value to be extracted, whereas for D2 we obtain \(-8.4 \pm 1\) MHz.

We note that because splitting due to hyperfine and quadrupolar coupling cannot be resolved, the chemical and structural compositions of a defect can in general not be determined. However, by comparing our findings with defects with known hyperfine structure, some types of defects can be excluded and information on the spin-density distributions can be gained. We restricted the comparison to four point defects addressed in h-BN EPR studies reported in the literature\(^{17,18,22-24}\). The experimental and simulated ODMR spectra for D1 are presented in Fig. 4a, and those of the four literature defects are depicted in Fig. 4b–e: the defects considered are a carbon atom substituting a nitrogen atom and hyperfine-coupled to three nearest-neighbour boron atoms (CTBC, Fig. 4b), a boron vacancy (VTBC, Fig. 4c) and two nitrogen vacancies (VTBC, Fig. 4d,e). For the VTBC defects, hyperfine features for two different configurations were measured: unpaired electrons coupled to either three nearest-neighbour B nuclei or just one B nucleus. These are referred to as the three-boron-centre nitrogen vacancy (VTBC\(^{\text{obc}} \), Fig. 4d) and the one-boron-centre nitrogen vacancy (VTBC\(^{\text{obc}} \), Fig. 4e), respectively\(^{17,18,22-24}\).

For the VTBC\(^{\text{obc}} \), oxidative damage typically forces the electron to interact only with a single B nucleus. In all cases, the g-factor is close to 2. This is in agreement with our measurement, within experimental error. In contrast, the hyperfine coupling (hfc) parameters vary significantly. For instance, VTBC\(^{\text{obc}} \) exhibits an axially symmetric hfc tensor with \( A_{\parallel} = 247.8/326.2 \text{MHz} \), whereas VTBC\(^{\text{obc}} \) shows two sets of values: \( A_{\parallel} = 112/127.8 \) and 18.4/22 MHz. These parameters were used in the spin Hamiltonian to compare simulations with the experimental results. The simulations are shown in Fig. 4d,e, neglecting the quadrupolar moment and assuming that only the spin-conserving, \( \Delta m = 0 \), transitions are allowed for electron-spin resonance. The line shape of the ODMR resonance depends on the number \( N \) of nuclei (with identical hfc parameter \( A \)) to which the electron is coupled. This leads to \( 2N+1 \) equally spaced lines in the spectrum. For instance, the coupling of a VTBC\(^{\text{obc}} \) with \( 11^\text{B} \) (\( I = 3/2 \)) generates 19 lines in the spectrum, in contrast to only 4 for the coupling of a VTBC\(^{\text{obc}} \) with \( 11^\text{B} \) \( (I = 3/2) \), as seen in the simulations for the different defects in Fig. 4d,e. We assumed that there is only hfc between the electron spin and \( 11^\text{B} \), and that the magnetic field is aligned along the \( A_{\parallel} \) direction. The simulation and the observed spectrum show a clear deviation (300 MHz) in comparison with the simulated spectrum of VTBC\(^{\text{obc}} \). The VTBC\(^{\text{obc}} \) simulation also deviates with an overall linewidth factor greater than 5. The EPR-measured\(^{12} \) hfc of a VTBC\(^{\text{obc}} \) is close to our observation (Fig. 4c). However, the neighbouring boron atoms give rise to only a few hyperfine lines and hence a clearly structured ODMR resonance line, not seen in our data. The carbon-related nitrogen vacancy CTBC\(^{\text{obc}} \) is the defect that has shown the smallest hfc in h-BN so far\(^{18} \). However, even for this, the hfc experimentally found for this defect type is too large to explain our experimental findings. Hence, we conclude that our measured ODMR spectra coincide with none of those reported so far\(^{12,14} \). In particular, the electron spin must possess an in-plane spin density that is significantly smaller than has previously been observed.

To gain further insight into the unpaired electron wave function, we next explored the ODMR linewidth dependence on the applied magnetic field tilt angle \( \theta \). The hyperfine tensor \( A \) can be parameterized as \( A(\theta) = A_{\parallel} + \Delta A \cos^2(\theta) \) with \( \Delta A = A_{\perp} - A_{\parallel} \). Figure 4f shows a comparison of the experimental ODMR data for two different angles with the corresponding simulations. A 30% decrease in linewidth from 41.3 to 31.2 MHz is observed when \( \theta \) is changed from 0 to 40°. The best fit to the data is obtained for the ratio \( A_{\parallel}/A_{\perp} = 10.6 \). Such a ratio is typical for unpaired electrons that reside in an extended \( z \)-orbital and are weakly coupled to

![Fig. 3 | ODMR and field dependence of the central frequency. a. Normalized ODMR contrast for MW sweeps of defect D1 at different external magnetic field values. The in-plane magnetic field values are indicated in each spectrum. b. ODMR spectrum for defect D2 recorded at a magnetic field of 288 G. The solid line is the Gaussian fit to the data. The FWHM of the resonance is 35 MHz. c, d, Central frequency of the ODMR envelopes for defects D1 (blue) and D2 (red) as a function of the in-plane magnetic field \( B_{\parallel} \) in the ranges 0–50 G (c) and 0–300 G (d).](https://example.com/fig3.png)
in-plane nuclear spins. Based on these hfc parameters, we derived an optimum fit of the ODMR line (Fig. 4a) assuming a homogeneous linewidth of ~7 MHz (Extended Data Fig. 8).

Recently, evidence has been accumulating that suggests such a defect structure consists of additive carbon impurity atoms, possibly introduced during exfoliation/thermal annealing. We carried out DFT calculations that revealed that the hyperfine structure can display a narrow linewidth yet with unresolved hyperfine interaction (Supplementary Section 11), as seen experimentally. The electron density for a carbon substituting a boron atom in h-BN (CB; Supporting Information) was calculated to have a large out-of-plane component, as concluded from the data shown in Fig. 4f and the small ZFS observed in our experiments. The hfc derived from the DFT calculations of CB (see Supplementary Section 11 for details) shows very good agreement with the ODMR spectrum (Fig. 4a). Further, we found agreement with the emission spectra obtained from the electronic structure calculations, which yielded the zero-phonon line energy ($E_{2B}$) in the range of 1.6–1.7 eV (Extended Data Fig. 9 and see the Supplementary Information for details).

Finally, we turned our attention towards the determination of the spin relaxation parameters. The best fit of the ODMR linewidth to our experimental data (Fig. 4a) yields a homogeneous linewidth of 7 MHz, equivalent to the lower limit of the spin coherence time $T_2 \approx 0.2 \mu s$. We note that this $T_2$ value is two orders of magnitude smaller than previously estimated for the NBVN point defect. This may be a result of residual neighbouring electron spins or residual unresolved hfc. From the power dependence of the ODMR contrast and linewidth, spin relaxation times can be extracted: $T_1 \approx 17 \pm 4 \mu s$ and $T_2^* = 57 \pm 10$ ns for D1, and $T_1 \approx 13 \pm 3 \mu s$ and $T_2^* = 41 \pm 3$ ns for D2 (Fig. 4g and Supplementary Section 6). The value of $T_1$ is compatible with the estimate deduced from pulsed ODMR in Fig. 2 (also see Extended Data Fig. 10). Room temperature (RT) ODMR was
not seen, possibly due to the rapid temperature dependence of $T_1$ (ref. 19). We note that similar $T_1$ values have been found previously in EPR studies on carbon-related defects in h-BN20.

In conclusion, we have studied isolated optical emitters embedded in h-BN that exhibit ODMR and ZFS of less than 10 MHz. Its angular dependence points to an unpaired, out-of-plane delocalized $\pi$-orbital electron, likely originating from substitutional impurity atoms. Although in-plane spin defects may also exhibit ODMR, the inevitable large hfc will result in broad spectral lines, making them potentially challenging to observe. Hence, ODMR studies on singular spin-bearing defects in h-BN provide selectivity for emitters that are only weakly coupled to the in-plane nuclear spins and that possess significant out-of-plane spin density, caused, for instance, by impurity atoms. Controlling the adsorption or intercalation of such impurity atoms may be an interesting recipe to synthesize, study and manipulate these 2D spin-bearing defects with intriguing physics.

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/s41563-021-00979-4.

Received: 20 July 2019; Accepted: 1 March 2021; Published online: 6 May 2021

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Methods
Sample preparation. We used single-crystal h-BN, which has become the benchmark for high-quality h-BN in research. Details relating to our sample and annealing methods are provided in the Supplementary Information. A SiO$_2$ substrate with dimensions of ~1 mm $\times$ 1 mm was marked into nine segments using a diamond blade. It was then thoroughly cleaned in a peroxymonosulfuric acid (H$_2$O$_2$/H$_2$SO$_4$) solution for 2 h, repeatedly rinsed in deionized water and dried in pure N$_2$ gas. The substrate was exposed to air plasma for 10 min. Bulk single-crystal h-BN was exfoliated using the Scotch Tape method and thin flakes were placed onto the SiO$_2$. No organic solution was used after exfoliation. The substrate and flakes were again exposed to air plasma for 10 min and annealed at 850 °C for 1 h.

ODMR. Numerous flakes were characterized in a RT confocal set-up to search for QEAs. These were tested for a magnetic field-dependent optical response. Photoemission was detected using two avalanche photo diodes in a Hanbury Brown and Twiss configuration, and the PL was collected with a spectrometer. After identifying a stable paramagnetic QE, a copper wire was placed in close proximity to the h-BN flake hosting this target QE. The sample was then cooled in a helium cryostat for further characterization. ODMR was generated using a Rohde & Schwarz SMIQ 03b MW source and a Mini Circuits ZHL-42W+ amplifier.

Data availability
Source data are provided with this paper. Any further data are available from the corresponding author upon request.

Code availability
All fits to the data were made with the commercial plotting software Origin. The fit functions and Matlab code (Easy spin) used for simulating the hyperfine coupling parameters can be made available upon request. The first-principles calculations were carried out using a stable non-free ab initio plane-wave supercell software package.

Acknowledgements
We acknowledge support from the Max Planck Society, the EU through project ASTERIQS and the ERC grant SMeI. (Grant No. 742610) as well as the DFG, J.H.S. acknowledges financial support from the EU Graphene Flagship Core 3 Project. A.G. acknowledges support from the National Office of Research, Development and Innovation of Hungary (NKFIH) within the National Excellence Programme (Grant No. 123213), and from the National Quantum Technology Program (Grant No. 2017-1.2.1-SDK-2017-00001), and from the Ministry of Innovation and Technology of Hungary within the National Quantum Informatics Laboratory. The growth of h-BN crystals was sponsored by the Elemental Strategy Initiative conducted by MEXT, Japan (Grant No. JP17H00354 and the CREST, Grant No. JPMJCR1715). The work at DGIST was supported by the Basic Science Research Program (NRF-2018R1D1A1A09084220) through the National Research Foundation of Korea (NRF), and also by the DGIST R&D Program (20-CoE-NT-01), funded by the Korea Ministry of Science and ICT. We thank M. W. Doherty for fruitful discussions.

Author contributions
All authors contributed to the paper. N.C., A.M., J.G. and Y.-C.C. contributed equally. N.C., J.W. and A.F. conceived and designed the experiments. T.T. and K.W. grew the single-crystal h-BN material. Y.K. and N.C. prepared the h-BN samples. A.D. fabricated the NV$^-$-implanted diamond. N.C., J.W. and A.G. performed all measurements. P.A. and A.G. carried out the first-principles calculations and analysed the results. N.C., A.M., D.B.R.D. and J.W. analysed the data. J.W. and J.H.S. supervised the project. N.C. and A.M. wrote the manuscript with input from J.W., D.B.R.D., A.F., A.G. and J.H.S.

Competing interests
The authors declare no competing interests.

Additional information
Extended data is available for this paper at https://doi.org/10.1038/s41563-021-00979-4. The online version contains supplementary material available at https://doi.org/10.1038/s41563-021-00979-4. Correspondence and requests for materials should be addressed to A.M. or D.B.R.D.

Peer review information Nature Materials thanks the anonymous reviewers for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Confocal scans and identification. a. The top panel displays spatial dependence of the emission for laser excitation at 633 nm wavelength. The bottom panel shows a confocal scan using low power excitation at 532 nm wavelength. It reveals that the emitter is located within the h-BN flake. The white scale bar corresponds to one micron. b. High resolution confocal scan of the emitter for 633 nm excitation. The emitter is less localized in comparison to the spread of another non-paramagnetic single emitter, (black bars represents the spatial extension in x and y profiles). For 35µW power of 633 nm laser excitation. The white scale bar corresponds to 400 nm.
Extended Data Fig. 2 | Optical polarization and auto-correlation. (Top) The background uncorrected data for \( g^2(t) \) is shown for defects D1 and D2. (Bottom) The photoluminescence intensity of defect D1 (left) and defect D2 (right) as a function of the linear polarization orientation of the incident laser light. The data points (grey dots) were recorded at 8.5 K. The solid lines are fits to the data sets as described in the text.
Extended Data Fig. 3 | Phonon map for emitters D1, D2, D3. Normalized PL intensity as a function of the energy detuning away from the zero phonon line (ZPL). This graph illustrates the importance of phonon coupling for emitter D1 (blue), D2 (red) and D3 (green) for 633 nm laser light excitation.
Extended Data Fig. 4 | Non-paramagnetic emitter \( g^2(t) \) and room temperature PL. a, Auto-correlation function \( g^2(t) \) of a non-paramagnetic emitter with a similar emission wave length as the paramagnetic emitters discussed in the main text. b, The emission spectrum of this non-paramagnetic emitter.
Extended Data Fig. 5 | Field characterization using NV centers in diamond. a, NV ODMR measurements. b, Illustration of the angle between the magnetic field and the optical axis of the h-BN QE. c, A schematic of the translation of the magnetic field angle relative to the NV center. d, Magnetic field angle calibration from NV to h-BN emitter. e, The h-BN QE ODMR peak frequency as a function of the angle. The grey values refer to the magnetic field strength.
Extended Data Fig. 6 | Kinetics of the electronic states. a, Energy level diagram used for modeling. b, Fluorescence autocorrelation measurement for longer time scales. The solid curve is a four parameter fit. The extracted time scale is indicated on the curve. c, Fluorescence intensity dependence on the excitation laser power. The arrow indicates the laser power for which most measurements were carried out. Otherwise the power level is stated. The solid curve is a parabolic fit, the dashed line represents the linear component of the fit. d, Record of the PL intensity as a function of the delay time the two excitation sequences shown in the inset. In comparison with the behavior of the PL intensity during sequence 1, the fluorescence saturates at a lower value for simultaneous excitation of laser and MW radiation (sequence 2). This suggests shelving into a long lived metastable state during the initial laser excitation pulse in sequence 1. e, Fluorescence recovery curves for the different pulse sequences shown in the inset. f, Same as (e) but for longer delay times. The data indicate a lifetime of the long lived (ll) metastable state greater than 20 ms. g, The re-pump process is probed with the sequences shown in the inset. During sequence 2 the population is pumped into the ll state by the initial combined MW and laser pulse and subsequently repumped into state e2 during the probe laser pulse of variable width before the final fixed readout pulse. In contrast, during sequence 1 the population is not pumped into the ll state. The recovery of the ground state population due to the laser re-pump mechanism in the absence of MW is evident. The solid line is a simulation based on the energy level diagram model.
Extended Data Fig. 7 | Hyperfine decomposition of the ODMR for different microwave power levels and $T_2^*$ calculation. a, Power dependent measurements for defect D1 at a fixed magnetic field of 155G. b, Lorentzian fits to the second (red) and third peak of the decomposed ODMR line shape. c, Lifetime $T_2^*$ extracted from the data in panel (a) assuming coupling to a one boron center. The outermost signal components are not considered for the estimation of $T_2^*$ as they are too weak. Hence, the lifetime is calculated using only the second (red) and third (orange) peak 2 (red) and 3 (orange) in panel (a). d, Estimates of $T_2^*$ for defect D2.
Extended Data Fig. 8 | Calculated Hyperfine couplings and ODMR. Calculation of the ODMR lineshape for defect D1 based on the hyperfine coupling calculated from the point defects: a, CB, b, CB(+)-CN-O-DAP-2, c, CB(O)-CN(-)-DAP-4, d, C2CN trimer (see table 2.1, 2.2, 2.3 and 2.4). Since NMR active carbon is rare (13C, 1.1%), only boron (11B, 80.1%) and nitrogen (14N, 99.6%) are assumed to contribute to the line broadening. The calculations are given as stick spectrum, that is only spectral components are shown but not natural linewidth is assumed.
Extended Data Fig. 9 | Photoluminescence (PL) of defect candidates-DFT. Photoluminescence (PL) of defect candidates that could indicate the experimentally isolated optical emitter D1: a, Comparison of PL spectra, b, comparison of phonon sidebands (PSBs).
Extended Data Fig. 10 | Estimate of lifetime $T_1$. a, Pulsed ODMR sequence. A 500ns MW pulse is applied after simultaneous laser and MW excitation (sequence 1) or only laser pulse excitation (sequence 2). The time delay between initial excitation and the readout pulse is varied. Each sequence is repeated 20,000 times ($N = 20k$) for every measurement frequency in order to enhance the signal-to-noise ratio of the ODMR. b, Simulation for the ODMR contrast as a function of the delay time when assuming $T_1$ approaches $\infty$. The ODMR contrast increases with delay time. c, Experimental ODMR data for both measurement sequences. Simulation assuming $T_1 = \infty$ is shown in the right top indicate increasing contrast with time delay. The extracted time decay constant is plotted next to each curve. d, Simulation of the ODMR contrast as a function of time delay assuming $T_1 = 10 \mu s$. The time decay constant is comparable to what is observed in the data.