



# Ten-second electron-spin coherence in isotopically engineered diamond



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Solid-state spin defects are a promising platform for quantum networks. A key requirement is to combine long ground-state spin-coherence times with a coherent optical transition for spin-photon entanglement. Here, we investigate the spin and optical coherence of single nitrogen-vacancy (NV) centres in (111)-grown isotopically engineered diamond. Our diamond-growth process yields a precisely controlled <sup>13</sup>C concentration and low-ppb nitrogen concentrations. Combined with the mitigation of 50 Hz noise using a real-time feedforward scheme and tailored decoupling sequences, this enables record defect-electron-spin coherence times of  $T_2 = 6.8(1)$  ms for a Hahn echo and of  $T_2^{DD} = 11.2(8)$  s under dynamical decoupling. In addition, we observe coherent optical transitions with a near-lifetime-limited homogeneous linewidth of 16.9(4) MHz (222(3) MHz inhomogeneous broadening under 515nm illumination) and characterize the spectral diffusion dynamics. These results provide new avenues to investigate the incorporation of impurities in diamond and new opportunities for improved spin-qubit control for quantum networks and other quantum technologies.

Solid-state colour centres provide a promising platform for quantum networks and distributed quantum computing<sup>1–5</sup>, as well as for quantum sensing and simulation<sup>6–9</sup>. A single colour centre constitutes a natural quantum network node, with an electron spin that forms a spin-photon interface<sup>2,3,10</sup> that is coupled to nuclear spins, which form a quantum register<sup>4,11,12</sup>. Recent breakthroughs include metropolitan-scale entanglement generation<sup>13</sup> and the demonstration of increasingly advanced quantum network protocols<sup>14</sup>. A key requirement for a quantum network node is to combine long spin coherence for high-fidelity qubits<sup>5</sup> with a coherent optical transition for spin-photon entanglement<sup>15–19</sup>, both of which benefit from realizing pure host materials with reduced sources of electric and magnetic noise (Fig. 1a).

Long electron-spin coherence times have been shown for various platforms, including colour centres in diamond<sup>20,21</sup>, silicon-carbide<sup>22</sup>, and silicon<sup>23</sup>, and rare-earth ions<sup>24</sup>. Coherence times are typically extended by using pulse sequences, like dynamical decoupling, which protect from magnetic noise<sup>20,22</sup>, and/or via isotopic purification of the host material to a spinless isotope, reducing the spin-bath noise<sup>16,22,25,26</sup>. Despite this recent progress, it remains important to further understand, control, and mitigate noise sources and prolong spin-coherence times, while preserving the colour centre's coherent optical transitions.

Here, we investigate the optical and spin coherence of single nitrogen-vacancy (NV) centres in isotopically engineered, high-purity diamond. We demonstrate an electron-spin coherence up to 10 seconds, the longest reported for a solid-state electron-spin qubit, in combination with nearly lifetime-limited homogeneous optical linewidths.

First, we present homoepitaxial diamond growth directly on (111) substrates, with precise control of the <sup>13</sup>C concentration. (111)-oriented epilayers are attractive for various types of quantum devices. However, achieving both high crystallinity and chemical purity is often considered more challenging in (111) than in (100) growth<sup>27–29</sup>. Despite this, we find that our diamonds feature low strain and nitrogen concentrations in the low-ppb regime.

Second, we study NV centres in these diamonds (Fig. 1b). We first examine the NV electron-spin coherence for different <sup>13</sup>C concentrations. We find that at low <sup>13</sup>C concentrations the Ramsey-coherence time  $T_2^*$  and spin-echo time  $T_2^{\text{Hahn}}$  are limited by 50 Hz noise from the electrical grid. To mitigate this noise source we develop a feedforward compensation scheme, resulting in the longest Hahn-echo coherence time reported for colour centres ( $T_2^{\text{Hahn}} = 6.8(1)$  ms). While 50 Hz noise is commonly considered in, for example, atomic physics<sup>30,31</sup>, these results highlight its — perhaps

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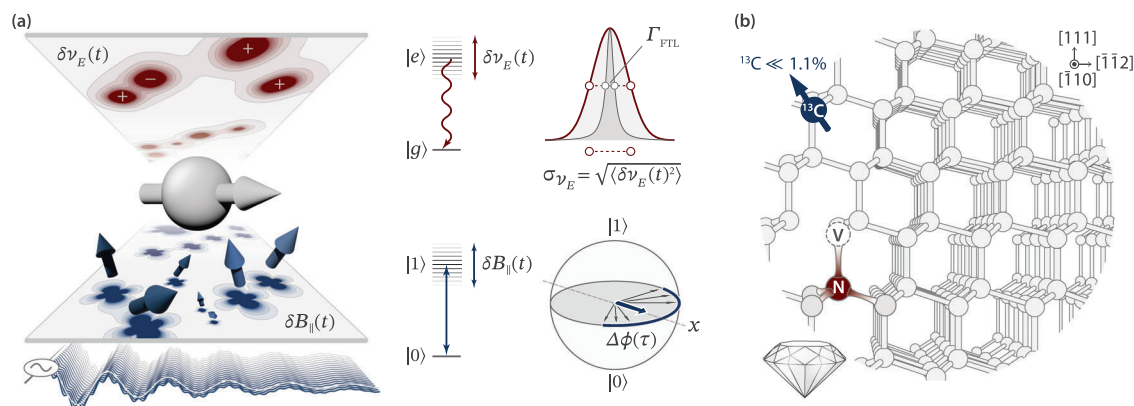


**Fig. 1 | Noise and coherence for colour centres.** **a** Colour centres are subject to electric field noise ( $\delta\nu_E(t)$ ) and magnetic field noise ( $\delta B_{||}(t)$ ), which affect their optical and spin properties. Electric field noise primarily originates from charges moving around in the host material, and causes frequency broadening and diffusion of the optical transitions. Magnetic field noise is commonly caused by electron and

nuclear spins in the host material, but external fields may also contribute, and is generally observed as decoherence of the ground-state spin qubit. **b** In this work we investigate the optical and spin coherence of nitrogen vacancy (NV) centres in high-purity (111)-grown diamond with varying  $^{13}\text{C}$  nuclear spin concentrations.

underappreciated — role in limiting colour-centre spin coherence. Using tailored decoupling sequences we extend the coherence time up to 11.2(8) s. Then, we further confirm the high material quality by showing near-lifetime-limited homogeneous optical linewidths. We probe the remaining spectral diffusion, which we show is predominantly laser induced, and develop a model that gives insight into the timescales at which spectral diffusion becomes significant.

The realized combination of isotopic engineering and record-long coherence times, while maintaining optical coherence, provides new opportunities towards high-fidelity quantum network nodes and towards precise sensing of signals originating from in and outside the diamond.

## Results

### Isotopic control in (111)-oriented homoepitaxial diamond

To engineer a high-quality host material for single NV centres, we employ Microwave-Plasma Chemical Vapour Deposition (MPCVD<sup>32</sup>) diamond growth with a controlled  $^{13}\text{C}$  concentration (Fig. 2a)<sup>33</sup>. Precise control of the  $^{13}\text{C}$  concentration is an enabling capability for quantum network experiments, due to the ability to trade longer electron-spin and nuclear-spin coherence times with the abundance of  $^{13}\text{C}$  spins for use as qubits<sup>25</sup>.

We chose to develop (111)-oriented diamond growth. While epitaxial growth on (111) surfaces is more challenging than (100) growth because of a tendency toward increased impurity incorporation<sup>27,28</sup> and a narrower growth window for suppressing twinning-driven morphological instabilities under high crystallization driving forces<sup>29</sup>, it offers several advantages. These include improved optical detection into single-mode fibres<sup>34</sup>, easier external magnetic field alignment with the [111] NV axis, and preferential alignment of NV centres that are formed during CVD growth along the surface-normal [111] direction<sup>35–38</sup>. Compared to cleaving (100)-grown diamonds, direct (111)-oriented growth enables large isotopically engineered layers and precise depth control of isotopes.

We grow homoepitaxial diamond layers on low-dislocation (111) high-pressure high-temperature (HPHT) diamond substrates (see Methods for details). This approach leverages the intrinsically high crystalline quality of HPHT diamond, which can yield a low overall dislocation density in homoepitaxial CVD layers<sup>39</sup>. Such epilayers are desirable for reducing possible impurity-incorporation variations associated with dislocation cores<sup>40</sup> and morphology-driven growth instabilities<sup>41</sup>.

The  $^{13}\text{C}$  concentration in the epilayer was controlled by mixing natural-abundance and  $^{12}\text{C}$ -enriched methane, each supplied through an independent mass flow controller (MFC) (Fig. 2a). We grow a calibration sample (named LD3) with a number of layers grown at varying flow ratios  $f_i = f_i/f_0$ , where  $f_0$  ( $f_i$ ) denotes the setpoint flow rate of  $^{12}\text{C}$ -enriched (natural-abundance) methane. The  $^{13}\text{C}$  isotope

concentration is subsequently analysed using Secondary Ion Mass Spectrometry (SIMS) (Fig. 2b). We define the  $^{13}\text{C}$  concentration as  $\chi = ^{13}\text{C} / (^{13}\text{C} + ^{12}\text{C})$  (see methods for the isotope concentration calibration procedure). Using only natural-abundance methane yields  $\chi_1 = 1.0937(86)\%$ , while only using  $^{12}\text{C}$ -enriched methane yields  $\chi_0 = 0.0013(3)\% = 13(3)$  ppm. When mixing the gases, the measured  $\chi_i$  (Fig. 2b) is reproduced by introducing an effective flow ratio  $\tilde{f}_i = f_i/f'_0$  such that

$$\chi_i = \frac{\chi_0 + \tilde{f}_i \chi_1}{1 + \tilde{f}_i}, \quad f'_0 = 1.023 f_0 + 0.036. \quad (1)$$

The lines in Fig. 2b are calculated using this calibrated effective-flow relation. With this definition the accessible  $^{13}\text{C}$  tuning range is 0.0139–1.0456%, which is constrained by the flow rate setting ranges of the MFCs.

### Nitrogen concentration

SIMS measurements of impurities other than  $^{13}\text{C}$  fall below the instrumental background ( $< 200$  ppb H,  $< 20$  ppb N,  $< 10$  ppb B). While the concentrations are low, the presence of these impurities may nevertheless be relevant for the spin or charge dynamics in the diamond, as well as for the local strain environment. Nitrogen is a particularly important contaminant due to its prevalence in air which inevitably leaks into the CVD reactor, and because it creates an electron-spin bath and a fluctuating charge environment that affect the spin and optical coherence of defect centres<sup>18,42</sup>. Therefore a more precise determination of the nitrogen concentration is desirable. Although sub-ppb nitrogen concentrations can be detected by ESR in bulk diamond samples of tens of milligrams<sup>43</sup>, the present samples, with approximately two orders of magnitude smaller mass, are expected to be sensitivity-limited for direct ESR measurements. In the following, we place an upper and lower bound on the nitrogen concentration by estimating the amount of nitrogen in the process gas and the efficiency of incorporation into the diamond.

We use the nitrogen incorporation efficiency  $\eta$  to relate the gas-phase  $\text{N}_2/\text{CH}_4$  ratio during growth to the solid-state nitrogen concentration in the diamond epilayer after growth (equivalently expressed as an N/C ratio)

$$[\text{N}]_{\text{ppb}} = \eta \frac{\dot{n}_{\text{N}_2}}{\dot{n}_{\text{CH}_4}} \times 10^9, \quad (2)$$

where  $\dot{n}_{\text{CH}_4}$  ( $\dot{n}_{\text{N}_2}$ ) is the methane (nitrogen) molar flow rate during growth.  $\dot{n}_{\text{CH}_4}$  is set by the MFC-regulated flow rate. We estimate  $\dot{n}_{\text{N}_2}$  by considering the vacuum leak rate of the reactor chamber, outgassing from chamber

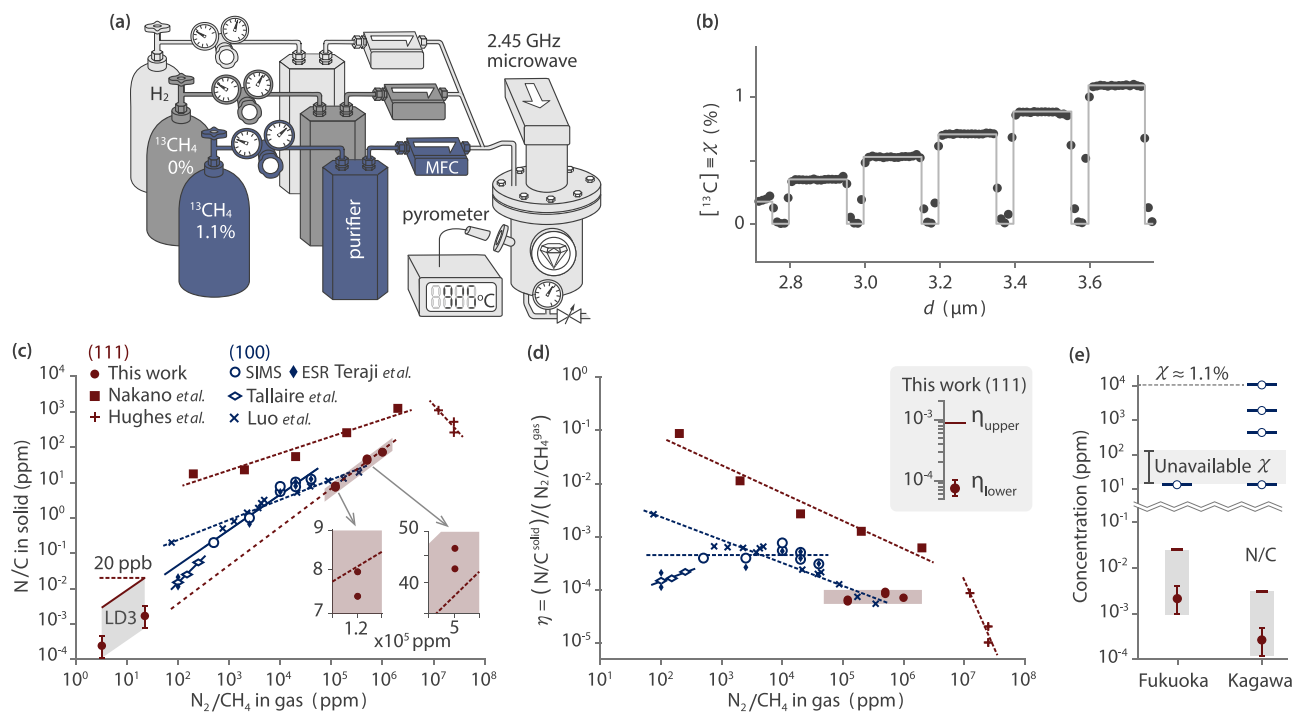


**Fig. 2 | Isotopically controlled diamond growth.** **a** Schematic of the MPCVD system. The ratio of natural-abundance and  $^{13}\text{C}$ -enriched methane flow, set by the mass-flow controllers (MFCs), controls the diamond  $^{13}\text{C}$  concentration  $\chi$ . **b** SIMS depth profile of the  $^{13}\text{C}$  calibration sample LD3 with multiple  $^{13}\text{C}$ -coded layers. **c** Solid-state N/C versus gas-phase  $\text{N}_2/\text{CH}_4$ , compiled from literature data and intentionally N-doped samples from this work. For the calibration sample LD3, the horizontal dashed line indicates the SIMS detection limit, while the shaded region indicates the leak-limited N/C range estimated from the  $\dot{n}_{\text{N}_2}/\dot{n}_{\text{CH}_4}$  ratio and the lower and upper bounds of the N-incorporation efficiency  $\eta$ . Insets show magnified views of the intentionally N-doped samples; light red shading denotes the known

day-to-day SIMS systematic uncertainty. **d**  $\eta = (\text{N/C})_{\text{solid}}/(\text{N}_2/\text{CH}_4)_{\text{gas}}$  extracted from the same dataset. The red shaded region indicates the range of  $\eta_{\text{lower}}$  estimated from the intentionally N-doped samples. The inset summarizes the bounds of  $\eta$  for this work. **e** Summary of the inferred N and  $^{13}\text{C}$  concentrations for the two NV samples studied (Fukuoka and Kagawa). Blue markers indicate  $\chi$  of each sample (Kagawa has 4 different layers). A small range of  $\chi$  is inaccessible because of the MFC tuning ranges. Red dots (lower bounds) and lines (upper bounds) indicate the estimated N concentration. Error bars indicate the propagated 95% confidence interval from both  $\eta_{\text{lower}}$  (from the intentionally N-doped samples) and the leak-derived  $\dot{n}_{\text{N}_2}$ .

materials, and impurity of the  $\text{CH}_4$  and  $\text{H}_2$  source gases (Suppl. Note 1). We find that air leaking into the reactor is the dominant source of nitrogen, with a nitrogen inflow rate of

$$\dot{n}_{\text{N}_2} = 4.0(1.6) \times 10^{-12} \text{ mol s}^{-1}. \quad (3)$$

Without purposeful nitrogen doping, [N] is therefore determined by the chosen methane flow rate  $\dot{n}_{\text{CH}_4}$  and the incorporation efficiency  $\eta$ .

We estimate  $\eta$  using lower and upper bounds. A lower bound is derived from SIMS measurements of three intentionally N-doped samples (red circles in Fig. 2c, d; see Methods for details), giving

$$\eta_{\text{lower}} = 7.5^{+2.6}_{-2.0} \times 10^{-5}. \quad (4)$$

We treat this value as a lower bound because  $\eta$  may increase at lower  $\text{N}_2/\text{CH}_4$  ratios<sup>44</sup>. For the upper bound, we use the  $^{13}\text{C}$  calibration sample LD3, which was grown without intentional nitrogen doping. Because  $\chi$  was controlled by changing  $\dot{n}_{\text{CH}_4}$ , the residual  $\dot{n}_{\text{N}_2}$  was effectively fixed, giving a range of  $\dot{n}_{\text{N}_2}/\dot{n}_{\text{CH}_4}$  ratios across the layer stack. Assuming that  $\eta$  remains approximately constant over this narrow gas-ratio range, we extract an upper bound on  $\eta$  by combining the largest ratio,  $\dot{n}_{\text{N}_2}/\dot{n}_{\text{CH}_4} \approx 23$  ppm, with the SIMS constraint of  $[\text{N}] < 20$  ppb:

$$\eta_{\text{upper}} < 8.9 \times 10^{-4}. \quad (5)$$

Figure 2c, d summarize our measurements of [N] and the extracted  $\eta$  together with literature results. The literature data include [N] extracted from SIMS<sup>28,45,46</sup> and ESR<sup>46–48</sup> for both (111) and (100) growth orientations (note that ESR only detects P1 centres,  $\sim 75\%$  of [N]<sup>46,48</sup>). The dashed lines

serve as guides to the eye, indicating overall trends in the log-log representation.

These lower and upper bounds constrain  $\eta$  to

$$0.55 \times 10^{-4} < \eta < 8.9 \times 10^{-4}, \quad (6)$$

which is lower than, or comparable to values reported for (100) growth. Using these bounds, we estimate [N] in the  $^{13}\text{C}$  calibration sample (LD3 in Fig. 2c), which is representative of our growth process. The range of  $\dot{n}_{\text{N}_2}/\dot{n}_{\text{CH}_4}$  ratios in the layer stack implies inferred layer-dependent [N] values of  $[\text{N}] = 0.2\text{--}2.3$  ppb from  $\eta_{\text{lower}}$  and  $2.9\text{--}20$  ppb from  $\eta_{\text{upper}}$ . For comparison, specifications of commercial {100} electronic-grade diamond are typically below 5 ppb [N] (Element Six), and ESR measurements on one such sample have reported P1 concentrations as low as  $0.07 \pm 0.02$  ppb<sup>43</sup>. Although the theoretical nitrogen-incorporation pathways differ substantially between (111)<sup>38</sup> and (100)<sup>49,50</sup> growth orientations, the resulting [N] is likely governed primarily by the local ratio of nitrogen- to carbon-containing reactive species at the growth surface. This interpretation is consistent with both our compilation of reported data in Fig. 2c,d and a broader literature survey<sup>44</sup>, which show no clear separation in [N] between (111) and (100) growth. The low [N] obtained here likely reflects a plasma-surface environment that is less favourable for N incorporation relative to carbon growth in comparison to other reported (111) growth conditions. Because this local N-to-C reactive-species ratio (represented here by atomic N and  $\text{CH}_3$ , respectively<sup>51</sup>) can vary spatially<sup>52</sup>, [N] is also likely to depend sensitively on the plasma-surface distance and sample-holder geometry, even under otherwise similar growth conditions. Finally, increasing the substrate temperature may further reduce [N], even in (111) growth<sup>53</sup>.

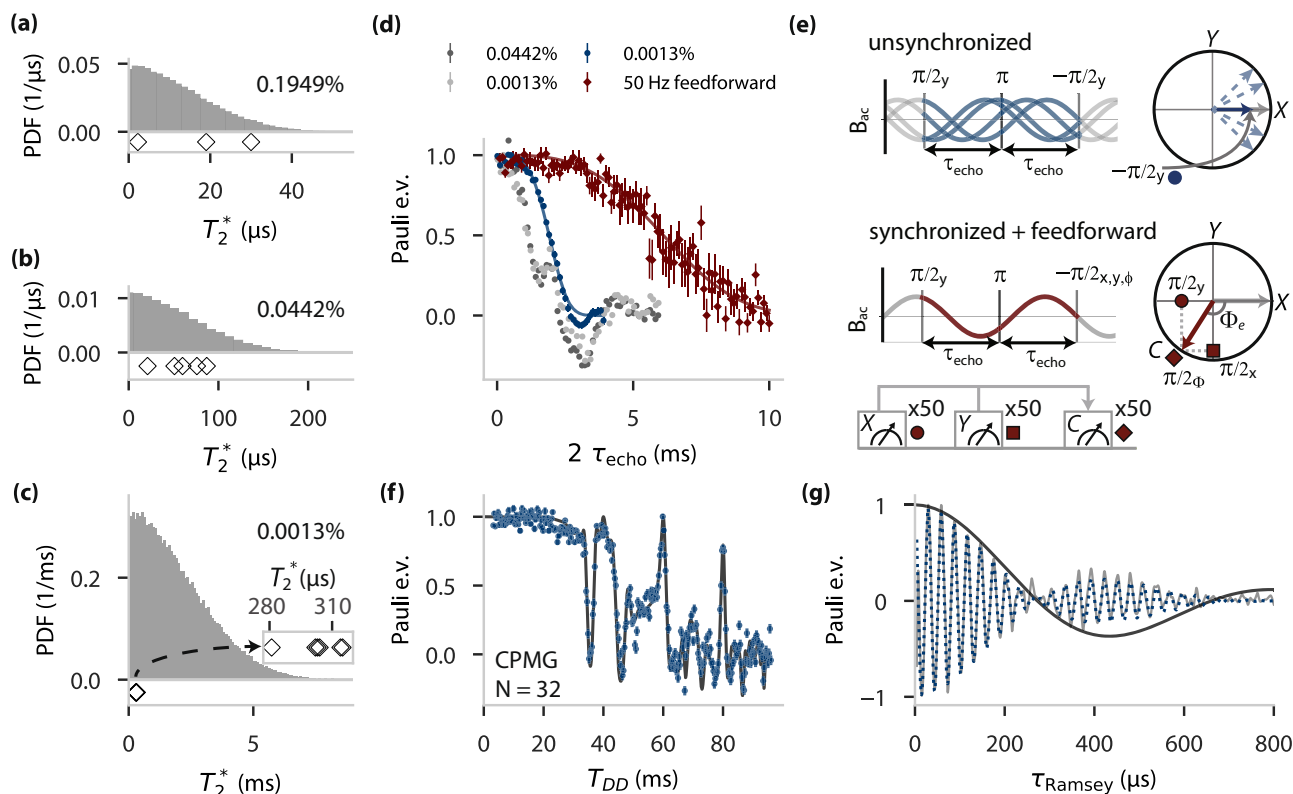


**Fig. 3 | Spin coherence in isotopically purified diamond.** **a–c** Expected  $T_2^*$  distribution (grey histogram) and measured  $T_2^*$  times (diamonds) for NV centres at  $\chi = 0.1949\%$  (**a**, sample Kagawa, 3 measurements),  $\chi = 0.0442\%$  (**b**, Kagawa, 5 measurements), and  $\chi = 0.0013\%$  (**c**, Fukuoka, 6 measurements). At the highest degree of purification,  $T_2^*$  is limited to roughly  $300 \mu\text{s}$ . An inset is used in (**c**) to more clearly show the  $T_2^*$  times. **d** Hahn-echo data showing  $T_2^{\text{Hahn}} \approx 2 \text{ ms}$  in Kagawa (grey) and in Fukuoka in a SIL NV (blue). We attribute the difference in data to the use of a different measurement setup. Using the 50 Hz noise mitigation scheme shown in (**e**),  $T_2^{\text{Hahn}}$  is prolonged to  $6.8(1) \text{ ms}$  (red). **e** When the Hahn-echo sequence in *unsynchronized*, external ac-fields cause dephasing. By triggering the start of the echo sequence with a clock that is *synchronized* to the mains frequency ( $\sim 50 \text{ Hz}$ ),

interference from the mains-frequency ac-fields can be measured and compensated using a feedforward scheme. We measure  $\langle X \rangle$  and  $\langle Y \rangle$  with 50 shots to estimate  $\Phi_e$ , and then measure the corrected Pauli observable  $\langle C \rangle$  along the axis defined by  $\Phi_e$ . Errorbars are derived from the standard deviation over 12 measurements of 50 shots each. **f** DD spectroscopy probing the mains interference. A model assuming 50 Hz ac fields (solid black line) was fit to the data, matching the measurements well. Note the revivals for 20, 40, 60 and 80 ms. **g** Ramsey signal of a SIL NV at  $\chi = 0.0013\%$ . We compare the data (grey line) to a fit using a product of two cosines and gaussian exponential decay (dashed blue line,  $T_2^* = 383.4(7) \mu\text{s}$ ), and to a prediction of the coherence envelope (dark grey line) by the 50 Hz noise model.

## NV-centre samples

Next, we investigate the spin and optical properties of NV centres in two diamond samples (Fig. 2e).

Sample *Fukuoka* was grown at the lowest possible  $^{13}\text{C}$  concentration  $\chi = 0.0013(3)\%$ . Based on the  $\text{CH}_4$  flow rate (0.19 sccm) the lower-bound  $\eta$  gives an estimated lower-bound  $[\text{N}]$  of 1–4 ppb, while our upper-bound estimate is  $[\text{N}] < 26 \text{ ppb}$ . Single NV centres are present in the diamond as-grown and are preferentially aligned along the surface-normal  $[111]$  direction (confirmed by single-NV ESR measurements), as is characteristic of  $(111)$  growth<sup>35–38</sup>. Solid-Immersion Lenses (SILs) were fabricated to enhance the collection efficiency<sup>54</sup> and striplines were fabricated for MW control of the NV centre electron spin (see methods for fabrication details).

Sample *Kagawa* features four different layers, each  $\sim 10 \mu\text{m}$  thick, with  $\chi = [0.0442, 1.0937, 0.1949, 0.0013]\%$  respectively. This allows us to investigate NV centres at different  $\chi$  on the same substrate, under as similar growth conditions as possible. A higher  $\text{CH}_4$  flow rate (1.53 sccm) results in a lower inferred  $[\text{N}]$ , with a lower-bound estimate of 0.1–0.5 ppb, and an upper-bound estimate of  $[\text{N}] < 3.2 \text{ ppb}$ . No naturally occurring NV centres were detected in the sample, consistent with the lower  $[\text{N}]$  estimate than *Fukuoka*. Instead, NV centres were created by 2 MeV electron irradiation (fluence  $5 \times 10^{11} \text{ cm}^{-2}$ ), followed by annealing at  $1000^\circ\text{C}$  for 2 h in vacuum.

## Electron-spin coherence

We investigate the electron-spin coherence of single NV centres and probe the noise sources that limit the Ramsey ( $T_2^*$ ), Hahn-echo ( $T_2^{\text{Hahn}}$ ), and CPMG dynamical decoupling (DD,  $T_2^{\text{DD}}$ ) coherence times. When the  $^{13}\text{C}$  spin bath is the limiting cause of dephasing, the coherence times are expected to scale with the  $^{13}\text{C}$  concentration  $\chi$  according to  $T_2^* \propto \chi^{-1.55}$  and  $T_2^{\text{Hahn}} \propto \chi^{-1.56}$ .

First, the electron-spin  $T_2^*$  of NV centres at three different  $\chi$  was measured (Fig. 3a–c). For a given  $\chi$ ,  $T_2^*$  for different NV centres will follow a probability distribution due to the random configuration of the nuclear spin environment of each NV centre (assuming that no other noise sources are present). We simulate the expected distribution (details in Suppl. Note 2).  $T_2^*$  follows the simulated distribution well for  $\chi = 0.1949\%$  and  $\chi = 0.0442\%$ , but for the lowest concentration  $\chi = 0.0013\%$ , the measurements are clustered around  $T_2^* \approx 300 \mu\text{s}$ . The clustering, and  $T_2^*$  being shorter than expected ( $\sim 2 \text{ ms}$ ), together indicate that another noise source limits  $T_2^*$ . We rule out that an electron-spin bath, arising from P1 centres or vacancy complexes, limits  $T_2^*$ . Such a bath would result in a similarly (half-normal) shaped  $T_2^*$  distribution, and thus is unlikely to yield the clustering around a non-zero  $T_2^*$ . Nevertheless the measurements of  $T_2^*$  place an upper bound of  $\sim 21 \text{ ppb}$  on the electron-spin bath concentration (Suppl. Note 3).

Hahn-echo measurements showed  $T_2^{\text{Hahn}}$  times that were limited to  $\sim 2 \text{ ms}$  (Fig. 3d), which is shorter than expected from just the  $^{13}\text{C}$  bath (up to  $\sim 800 \text{ ms}$  for  $\chi = 0.0013\%$ <sup>56</sup>). Other studies on defect centres across different

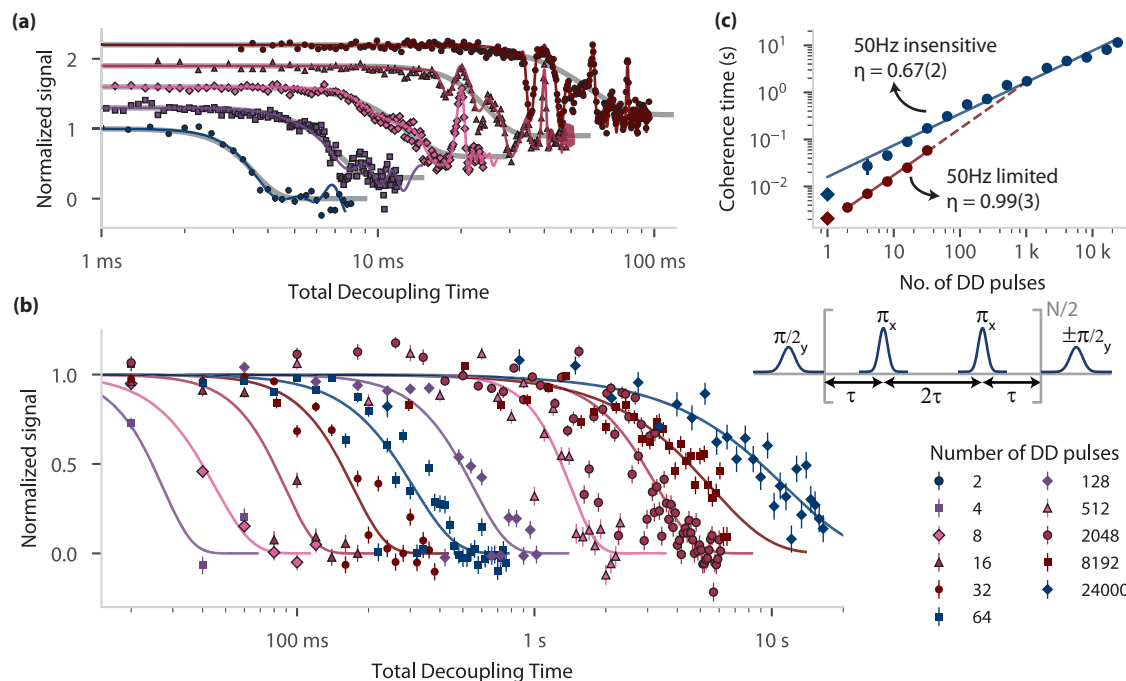


**Fig. 4 | Dynamical decoupling.** **a** High resolution CPMG data at  $\chi = 0.0013\%$ , showing the collapses and revivals due to 50 Hz noise. Data are each offset by 0.3 on the y-axis for clarity. Coloured solid lines are from the 50 Hz model fit ( $N = 32$  data also shown in Fig. 3f). The grey curves are decay fits of the data (Eq. (10)) when the signal revivals are ignored. **b** CPMG data with more decoupling pulses. Up to  $N = 2048$ , 50 Hz noise was mitigated by choosing interpulse delays  $\tau$  such that  $T_{DD}$  is a multiple of 20 ms. Data and fits are rescaled by the fit parameter  $A$ . Inset:

Dynamical decoupling sequence. **c** Extracted coherence time versus the number of decoupling pulses. The coherence time follows  $T_2^{DD} = T_0 N^\eta$ . In the case where 50 Hz noise is not mitigated by taking data at revival times (grey lines in **a**),  $T_2^{DD}$  scales linearly with  $N$  (red line,  $T_0 = 1.8(1)$  ms). Mitigating 50 Hz noise results in  $\eta = 0.67(2)$  (fits from **b**, solid blue line,  $T_0 = 16(2)$  ms). Diamond data points at  $N = 1$  are the  $T_2^{Hahn}$  times with (blue) and without (red) feedforward correction for 50 Hz noise.

isotopically purified materials (diamond, SiC, and Si) have reported similar  $T_2^{Hahn}$  times (in the 1.5–2.5 ms range)<sup>21–23,26,58,59</sup>. We next show that magnetic fields synchronous to the  $\sim 50$  Hz frequency of the mains electricity grid are limiting our observed  $T_2^{Hahn}$  and  $T_2^*$ . We note that, although the effect of mains-frequency signals on qubit coherence is commonly considered in fields such as atomic physics<sup>30,31</sup>, the recurring observation of limited  $T_2^{Hahn}$  in the literature suggest that this might have been overlooked for colour centres. While one mitigation path is to physically reduce the 50 Hz interference, e.g., through shielding or moving equipment, here we choose instead to mitigate this noise through a qubit-based feed-forward scheme and tailored pulse sequences.

The mains interference can be described by an ac magnetic field along the z-axis of the NV

$$B_{ac}(t) = \sum_{i=1}^n B_i \cos(\omega_i(t - t_0) + \phi_i), \quad (7)$$

where the summation is performed over  $n$  frequency components at 50 Hz and higher harmonics ( $\omega_i$ ), each with amplitude  $B_i$  and relative phase  $\phi_i$ .  $t_0$  is the delay between the start of the pulse sequence ( $t = 0$ ) and a reference point in the mains waveform. In a spin echo, the electron spin picks up a phase  $\Phi_e$  according to

$$\Phi_e = \gamma_{NV} \left[ \int_0^\tau B_{ac}(t) dt - \int_\tau^{2\tau} B_{ac}(t) dt \right], \quad (8)$$

where  $\gamma_{NV}$  is the NV centre gyromagnetic ratio, and  $\tau$  is the echo time. The spin-echo measures  $\langle X \rangle = \cos \Phi_e$ .

The fact that 50 Hz interference is limiting the coherence can be seen by synchronizing the start of the spin echo to the mains frequency (Fig. 3e). When the sequence is unsynchronized,  $t_0$  is randomized every measurement repetition, causing a Bessel-like dephasing for  $\langle X \rangle$ . When instead

the sequence is synchronized, here achieved by triggering the spin-echo sequence at a positive zero-crossing of the 50 Hz mains waveform,  $t_0$  is fixed, causing  $\Phi_e$  to be deterministic.  $\langle X \rangle$  then oscillates coherently as a function of  $\tau$ , assuming that  $B_i$  and  $\phi_i$  are constant over the course of the measurement.

The synchronization allows a feedforward scheme that partly corrects the 50 Hz interference. For a specific spin-echo time  $\tau$ , we measure  $\langle X \rangle$  and  $\langle Y \rangle$  to estimate  $\Phi_e$ , with

$$\Phi_e = \arctan \frac{\langle Y \rangle}{\langle X \rangle}. \quad (9)$$

The 50 Hz interference is then corrected during subsequent echo experiments by shifting the phase of the final  $\pi/2$ -pulse by  $-\Phi_e$ , yielding a new expectation value for the spin coherence  $\langle C \rangle$ .

For a SIL NV at  $\chi = 0.0013\%$ ,  $\langle X \rangle$ ,  $\langle Y \rangle$  are each measured with 50 shots, and  $\langle C \rangle$  are each measured with 50 shots (Fig. 3d, red diamonds). The feed-forward  $\langle C \rangle$  data results in  $T_2^{Hahn} = 6.8(1)$  ms, the longest reported for a defect centre electron-spin qubit. The remaining decay and larger spread in  $\langle C \rangle$  are caused by fluctuations in the ac-field magnitudes  $B_i$  during the  $\Phi_e$  estimation, which occur on the time scale of a few seconds (Supplementary Note 5). The choice for the number of shots (20 ms each due to the 50 Hz synchronization) is a trade-off between the accuracy with which  $\Phi_e$  is determined and the time-scale of fluctuations in  $B_i$  that can be tracked.

We then performed CPMG dynamical decoupling to further extend the coherence time and probe the noise spectrum (Fig. 3f and Fig. 4a). These measurements were not synchronized to the mains frequency. The phase  $\Phi_{DD}$  picked up by the spin can be described by  $B_{ac}(t)$  and the filter function of the CPMG sequence  $F(\omega, N, \tau)$ , with  $N$  the number of  $\pi$ -pulses, and  $2\tau$  the delay between  $\pi$  pulses (see Supplementary Note 4 for details). This enables a fine-grained analysis of  $B_{ac}(t)$ . We fit a model of the mains interference to

**Table 1 | Parameters for the 50 Hz interference model**

| Frequency (Hz) | $B_{ac}$ (mG) | $\gamma_{NV}B_{ac}$ (kHz) | $\phi_{ac}$ (rad) |
|----------------|---------------|---------------------------|-------------------|
| 50             | 2.95(3)       | 8.28(8)                   | 0                 |
| 100            | 0.024         | 0.067                     | 3(5)              |
| 150            | 0.490(6)      | 1.37(2)                   | -1.77(3)          |
| 200            | 0.0065(8)     | 0.018(2)                  | -0.4(8)           |
| 250            | 0.046(2)      | 0.129(4)                  | 4.33(9)           |
| 300            | 0.010(2)      | 0.029(6)                  | 0(3)              |
| 350            | 0.0376(1)     | 0.105(3)                  | 0.9(3)            |
| 450            | 0.0409(1)     | 0.115(3)                  | -1.3(4)           |

The phase of the 50 Hz component is chosen to be 0. Both the extracted field amplitude  $B_{ac}$ , and the corresponding shift on the electron-spin frequency  $\gamma_{NV}B_{ac}$  are reported. The amplitude of the 100 Hz component is chosen to match the CPMG data at  $\tau = 2.5$  ms (Suppl. Note 4), where only 100 Hz noise is detected.

the CPMG data (up to  $N = 32$ ), assuming that  $B_{ac}(t)$  is described by 50 Hz harmonics up to 450 Hz, with  $B_i$  and  $\phi_i$  for each harmonic as a free parameter (Table 1). The model reproduces the data well, confirming that 50 Hz interference is the dominant noise source for CPMG measurements at these values for  $N$ .

The  $B_{ac}(t)$  model allows us to consider the effect of the 50 Hz interference on Ramsey measurements. We observe a decaying beating signal (Fig. 3g). A naive fit to a product of two cosines yields a decay time of  $T_2^* = 383.7(7) \mu\text{s}$ . However, a comparison to the predicted signal envelope, based on  $B_{ac}(t)$  obtained from the CPMG data (Table 1), indicates that the 50 Hz interference also is the external noise source that limits the Ramsey signal and  $T_2^*$  (Fig. 3c). Note that the predicted envelope is averaged over a range of amplitude fluctuations of  $B_{ac}(t)$ , as observed in the Hahn-echo data (Suppl. Note 5).

Finally, we investigate the coherence with an increasingly higher number of pulses  $N$  (Fig. 4b, complete dataset in Suppl. Note 6). To avoid the 50 Hz noise, we set the interpulse delays  $\tau$  to follow the revivals at  $T_{DD} = 2N\tau = k \times 20$  ms ( $k \in \mathbb{N}$ ) (Fig. 4a). These revivals occur because, for most  $\tau$  at these  $T_{DD}$ , the CPMG filter function  $F(\omega, N, \tau) = 0$  for 50 Hz harmonics (See Supplementary Note 4 for exact conditions). We fit the function

$$S(T_{DD}) = A \exp[-(T_{DD}/T_2^{DD})^n] \quad (10)$$

to the data, with  $A$ ,  $T_2^{DD}$ , and  $n$  free parameters. Increasing the number of decoupling pulses to 24,000 results in a coherence time of 11.2(8) s, the longest coherence time reported for a single electron spin in a solid<sup>20,22</sup>.

Further insight can be obtained by studying the scaling of the coherence time with the number of decoupling pulses  $T_2^{DD} = T_0 N^\eta$  (Fig. 4c). As a comparison, we first consider the hypothetical case in which the pulse sequence is not tailored to avoid 50 Hz noise, and a naive dephasing curve is fit to the initial coherence decay while ignoring (or missing) the sharp signal revivals (Fig. 4a, grey lines). The coherence time then scales linearly with pulse number  $\eta \approx 1$ . We note that this is conceptually similar to the initial collapse of the electron-spin coherence in a nuclear spin bath<sup>60</sup>. Taking data at the revival times, thereby circumventing the 50 Hz noise, we find longer coherence times, with a scaling  $\eta = 0.67(2)$ . Such a scaling is consistent with the  $\eta = 2/3$  scaling caused by a Lorentzian ( $\omega^{-2}$ ) noise spectral density, as would be caused by e.g., an electron-spin bath<sup>61–63</sup>.

The two observed scalings imply a crossover near  $N \approx 1000$ . This coincides with the highest detected 50 Hz harmonic of 450 Hz, which results in a dip at  $\tau = 1/(4f) \approx 556 \mu\text{s}$ . At  $N = 1000$ , this dip occurs at a total decoupling time of  $2N/4f = 1.1$  s, which indeed is at the crossover point. Thus, beyond  $N \approx 1000$  the 50 Hz interference is no longer the limiting noise source for the  $T_2^{DD}$ , and it is no longer necessary to follow the signal revivals. Separate scaling behaviour for low and high  $N$  is also reported in other

work<sup>22</sup>; 50 (or 60) Hz noise might have also played a role in those results (see Supplementary Note 7). Assuming that the 2/3-scaling corresponds to a [N]-limited coherence regime we can extrapolate a hypothetical  $T_2^{\text{Hahn}} = 16$  ms from the fit. Using the (NV ensemble) results from Bauch et al.<sup>64</sup> which relate  $T_2^{\text{Hahn}}$  and [N], yields [N]  $\approx 10$  ppb, which is in agreement with our estimated range for [N] (Fig. 2e).

### Optical properties

For applications that use the NV centre as a photon source, such as quantum networks, a coherent optical transition with limited spectral diffusion due to charge noise is required<sup>15,19</sup>. Here we investigate the same NV centre that was used for the CPMG measurements (Fig. 4), and show that our diamond growth yields good optical properties. Note that previous investigations of isotopically purified diamond observed increased spectral diffusion in comparison to natural abundance diamond, potentially caused by the presence of additional impurities<sup>25,59,65</sup>.

First, we determine the optical level structure via a Photo-Luminescence-Excitation (PLE) scan (Fig. 5a). A 515 nm repump pulse is followed by a probe pulse at frequency  $f_p$ , while microwaves are applied to mix the  $m_s = \pm 1$  and  $m_s = 0$  states. The spectrum (Fig. 5b) matches the known NV-centre level system and confirms that the NV is in a low lateral strain regime ( $V_E = 2.1$  GHz), comparable to values achieved in (100) CVD-grown and HPHT diamonds<sup>66,67</sup>.

Secondly, we determine the linewidth of the  $E_y$  optical transition. The lowest attainable (Fourier-limited) linewidth is set by the excited-state lifetime, but fluctuating charge traps arising from crystal defects can cause spectral diffusion, effectively broadening the optical transition. We use check-probe measurements to determine the linewidth<sup>68</sup> (Fig. 5c). First, a check pulse at frequency  $f_c$ , resonant with the  $E_y$  transition, checks the charge state and frequency resonance of the emitter, which are heralded by the number of check counts  $C$  exceeding a set threshold  $C_{th}$ . Subsequently, a detuned probe pulse at  $f_p$  probes the response of the emitter, yielding  $P$  probe counts. For  $f_p = f_c$ , this is similar to the common charge-resonance check used to prepare the charge state and frequency resonance of defect centres prior to experiments<sup>69,70</sup>. The measurement is repeated for different  $f_p$  to determine the linewidth. CW microwaves are applied during laser pulses to mix the ground state spin levels, to avoid spin pumping.

Setting no threshold ( $C_{th} = 0$ ) yields the inhomogeneous linewidth  $\gamma_i = 222(3)$  MHz (FWHM, Fig. 5d). This linewidth arises from charge scrambling induced by the repump pulse prior to each measurement. Thresholding at  $C_{th} = 30$  approaches the homogeneous line with FWHM  $\gamma_h = 16.9(4)$  MHz, close to the NV-centre lifetime-limited linewidth of  $\sim 13$  MHz and among the lowest values measured in literature (Fig. 5e)<sup>15,18</sup>. With charge-resonance checks, spin-selective excitation gives an average NV single-shot readout fidelity of 92.5%<sup>71</sup>. We note that this single-shot readout was also used to enable the long spin coherence measurements in the previous section.

To characterize the spectral stability after frequency resonance is heralded, we measure the diffusion rate during optical excitation (Fig. 5f)<sup>68,72</sup>. Here, two “check” blocks ( $C_1$  and  $C_2$ ) are separated by a diffusion period with duration  $\tau_d$ , during which a laser at  $f_{RO}$  is applied to induce diffusion. The check blocks consist of simultaneous application of lasers at  $f_{RO}$  and  $f_{SP}$  to continuously excite all ground-state spin states. A high number of  $C_1$  counts heralds frequency resonance of the emitter. When spectral diffusion occurs during the diffusion period, this will cause the subsequent number of  $C_2$  counts to be smaller. The time scale at which significant diffusion occurs while the readout laser is applied determines how many coherent photons the defect centre can emit before the frequency resonance has to be re-prepared. Both correlations forward in time and backward in time can be considered, by post-selecting  $C_2$  counts conditioned on  $C_1 > C_{th}$  ( $C_2[C_1 > C_{th}]$ ), and vice-versa ( $C_1[C_2 > C_{th}]$ ). This

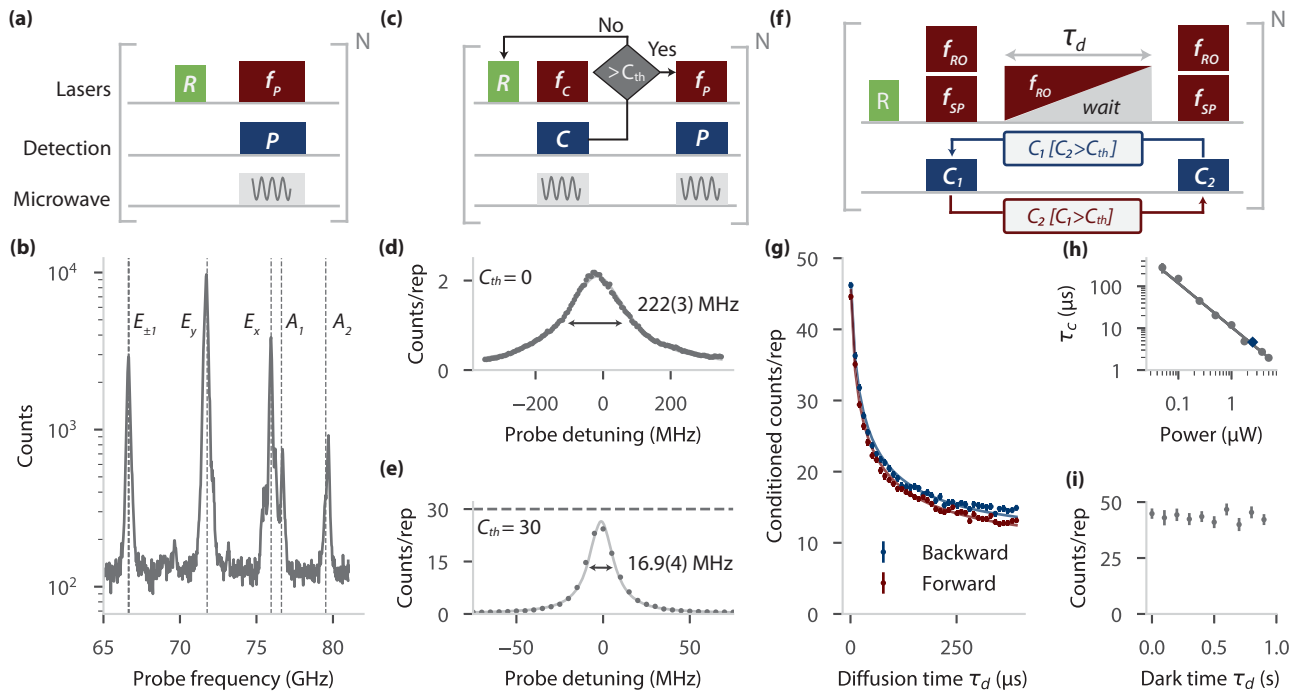


**Fig. 5 | Optical coherence and spectral diffusion.** **a** Diagram for a PLE scan. A repump laser pulse  $R$  at 515 nm ( $20 \mu W$ ,  $50 \mu s$ ) is followed by a *probe* pulse at frequency  $f_p$ , resulting in  $P$  detected photons. Microwaves at the  $m_s = \pm 1 \rightarrow m_s = 0$  transitions are applied during the probe pulse. **b** PLE scan showing a low-strain NV optical spectrum. **c** Check-Probe measurement scheme<sup>68</sup>.  $f_c$  is kept at the  $E_y$  transition, while the detuning  $f_c - f_p$  is varied using  $f_p$  (both pulses  $400 \mu s$  at  $1 nW$ ). Each  $f_p$  is measured  $N = 50000$  times. **d** Check-probe PLE of  $E_y$ . Without thresholding ( $C_{th} = 0$ ), the inhomogeneous linewidth is  $222(3)$  MHz (Lorentzian fit). **e** Using  $C_{th} = 30$  gives a homogeneous linewidth of  $16.9(4)$  MHz with a check success probability of  $1.4\%$ . **f** Spectral-diffusion measurement scheme<sup>68</sup>. Check blocks before

( $C_1$ ) and after ( $C_2$ ) the diffusion period of duration  $\tau_d$  consist of simultaneous application of pulses at  $E_y$  ( $f_{RO}$ ) and  $E_{\pm 1}$  ( $f_{SP}$ ) for  $100 \mu s$  at  $5 nW$ . We consider correlation forward and backward in time, by only selecting  $C_2$  counts for repetitions where  $C_1 > C_{th}$  and vice-versa. We set  $C_{th} = 40$ , giving selection probabilities in the  $2-4\%$  range ( $N = 40000$ ). **g** Data for a  $P = 2.5 \mu W$  diffusion pulse at  $f_{RO}$ . Solid lines are from a model assuming O-U diffusion of the spectral frequency (backward) and additionally frequency-dependent ionisation (forward). **h** The extrapolated diffusion timescale  $\tau_c$  scales as  $P_d^{-1.05(4)}$ . Blue diamond corresponds to data shown in (g). **i** In the dark, no significant diffusion is observed.

separates ionization, which occurs only in forward-time, and diffusion, which is a time-symmetric process<sup>68</sup>.

The data reveals a relatively time-symmetric decrease in photon counts, a signature of spectral diffusion dominating over ionisation (Fig. 5g). The photon counts tend to a non-zero value for long  $\tau_d$ , in contrast to recent experiments on the  $V_2$  defect in SiC where a decay to 0 counts was observed<sup>68</sup>. This is caused by a relatively small ratio of  $\gamma_i/\gamma_h$ ; The inhomogeneous linewidth limits the allowed frequency range of the spectral diffusion, so for smaller  $\gamma_i$  there remains a probability for the transition frequency to be close enough to  $f_{RO}$ , even for long  $\tau_d$ .

To account for this difference, we adapt previous models<sup>68</sup> by describing the spectral diffusion as an Ornstein-Uhlenbeck (O-U) process<sup>72</sup>, which describes a random walker confined to a harmonic potential. The model predicts the probability distribution  $P(f, \tau_d)$  of the transition frequency  $f$  over time. The dynamics are determined by two parameters: the diffusion parameter  $D$ , which is related to the timescale at which diffusion occurs, and the inhomogeneous linewidth  $\gamma_h$ , which determines the frequency range that the emitter can diffuse over. This model is chosen because it reproduces the Gaussian steady-state probability distribution for  $\tau_d \rightarrow \infty$ : the inhomogeneous spectral lineshape. In O-U diffusion,  $P(f, \tau_d)$  is a Gaussian distribution with a time-dependent variance

$$V(\tau_d) = \frac{\gamma_i^2}{8\ln(2)} \left( 1 - \exp \left[ \frac{-2D\tau_d}{\gamma_i^2/(8\ln(2))} \right] \right). \quad (11)$$

To relate  $D$  to a more easily interpretable physical quantity, we define  $\tau_c$  as the diffusion duration for which the FWHM of  $P(f, \tau_c)$  is equal to the homogeneous FWHM  $\gamma_h$ ; this sets a rough timescale at which the diffusion

significantly affects the optical linewidth.  $\tau_c$  and  $D$  are related via

$$\tau_c \approx \frac{\gamma_h^2}{16\ln(2)D}, \quad (12)$$

assuming  $\gamma_i^2/\gamma_h^2 \ll 1$ . Our model first calculates  $P(f, \tau_d)$ , which is then convolved with a Lorentzian of height  $C_0$  and linewidth  $\gamma_h$ , corresponding to the homogeneous response, to calculate the expected number of counts  $C(\tau_d)$ . We fix  $\gamma_h = 22$  MHz, which is set by the power broadening during the  $C_1$  and  $C_2$  pulses (see Supplementary Note 8 for more details).

We perform the diffusion measurement for a range of diffusion-pulse powers  $P_d$ . We perform a joint fit of the model to the backward-time correlation data for all  $P_d$ , fitting a single  $\gamma_h$ , with separate  $D$  and  $C_0$  for each  $P_d$  (Fig. 5g). This choice is motivated by the expectation that the inhomogeneous linewidth is determined by the number of fluctuating charge traps in the environment, which is expected to primarily depend on the photon energy, not the laser intensity. The model fits well to the data (reduced chi-squared  $\chi_r^2 = 2.1$ ), indicating that the assumption of a power-independent  $\gamma_i$  is reasonable (see Supplementary Note 8 for data at all powers). The diffusion timescale scales as  $\tau_c \propto P_d^{-1.05(4)}$  (Fig. 5h). At  $P_d = 15 nW$  (the saturation power when a  $1 \mu s$  pulse is applied), the fit predicts  $\tau_c > 0.8$  ms. Recent measurements on T-centres<sup>72</sup> and erbium atoms<sup>73</sup> in silicon show a similar scaling of spectral diffusion with  $P_d$ . Without an applied laser, no spectral diffusion is detected within one second (Fig. 5i). The fit inhomogeneous FWHM  $\gamma_{i,red} = 117(2)$  MHz from our model corresponds to the inhomogeneous linewidth due to the diffusion laser (near 637 nm). This is smaller than  $\gamma_i$  under 515 nm excitation ( $222(3)$  MHz, Fig. 5d), which is likely caused by the lower energy 637 nm light exciting a smaller number of

charge-trap species. Under 515 nm excitation we also observe diffusion timescales that are an order of magnitude faster at the same laser power, implying that  $\tau_c$  also has a photon-energy dependence.

To also model the forward-time data we consider the  $NV^- \rightarrow NV^0$  ionisation rate, which is caused by resonant  $f_{RO}$  laser light. We use the same diffusion model and parameters as for the backward-time fits. First of all, we adjust the model counts for an overall slight decrease (4%) in the forward-time counts for all  $P_{db}$  caused by ionisation during the  $C_1$  and  $C_2$  pulses. For ionisation during the diffusion period we develop an additional model, which takes into account that ionisation only occurs when the emitter is resonant with the diffusion laser. Because there is no closed-form solution for O-U diffusion with frequency-dependent ionisation, our model instead performs an asymptotic approximation of  $P(f, \tau_d)$ , assuming a  $\delta$ -function ionisation rate when  $f = f_{RO}$ <sup>74</sup> (see Suppl. Note 9).

The methods described here enable quantitative comparison between emitters in different diamonds. To put the obtained values in perspective, we perform the same spectral diffusion measurements on an NV centre that was previously used in remote entanglement experiments<sup>13</sup> (Suppl. Note 10), and is hosted in a non-isotopically-purified (100)-grown diamond (Element Six). We find a smaller  $\gamma_{i,red} = 42(9)$  MHz and values for  $D$  that are  $\sim 30$  times lower. This comparison to the state-of-the-art for (100)-oriented growth shows that, while our (111)-oriented growth results in stable lines with limited spectral diffusion, further improvements in material purity might be possible. Reducing the diffusion rate would, for example, enable more repetitions of remote entanglement protocols before requiring a new charge-resonance check, which can increase the overall entanglement rate. While detailed, broad laser frequency scans would be required to attribute the spectral diffusion to specific defect types, we hypothesize that in our (111)-grown sample, the spectral diffusion under 637 nm excitation is caused by photoionisation of single vacancies (GR1, photoionization at 1.67 eV<sup>75,76</sup>) and vacancy complexes that were incorporated in the CVD film during growth, or possibly by hydrogen-related defects (1.2 eV)<sup>77</sup>. The increase in diffusion during 515 nm excitation is likely caused by the remaining  $N_s$  centres, which are not efficiently ionised by 637 nm excitation (photoionization at 2.2 eV<sup>18,75,77</sup>).

## Discussion

We have presented NV centres in isotopically engineered diamond with electron-spin coherence times up to 11.2(8) s, in combination with stable optical transitions. These results are made possible by: (1) a high-purity, low-strain, (111)-oriented diamond growth process with a precisely controlled  $^{13}C$  concentration; (2) spin-echo sequences that characterize and mitigate 50-Hz related noise, and (3) quantitative methods to characterize the spectral diffusion.

Our results open several new avenues. The quantitative analysis of spectral diffusion enables systematic comparisons of optical coherence between different emitters and material platforms, and the long electron-spin coherence can be used to precisely probe spin impurities. Together with future investigations into the nature of the impurities responsible for the spectral diffusion, for example by measuring frequency dependencies<sup>18</sup>, or by directly probing dark electron spins<sup>7,78</sup>, our methods can inform further improvements in high-purity material growth.

In addition, our results provide further insight into the electron-spin coherence and the degree of isotopic purification that still yields practical benefit. When coherence starts to be limited by factors other than the nuclear spin bath, further isotopic purification primarily reduces the number of  $^{13}C$  spins that can be used as qubits, for example as memory qubits<sup>25</sup>. In addition, the initially observed limit to  $T_2^{Hahn}$  and  $T_2^*$  values for low  $^{13}C$  concentrations, and the consistency with which these are found in the literature<sup>21–23,26,58,59</sup>, highlight the importance of characterizing and mitigating 50 Hz related noise. Further improvements in spin coherence will likely be based on isotopic purification in combination with reducing external magnetic noise (e.g., by magnetic shielding) and spin impurities in the diamond.

An important consideration is the source of the 50 Hz noise. The noise either couples to the NV centre through currents that run through the drive line (e.g., because of a ground loop), or ambiently, for example, by magnetic fields that originate from transformers in lab equipment. To eliminate the first source, an inner-outer DC block was used in the signal chain, which prolonged the coherence somewhat, though not beyond 3 ms. This implies that a significant portion of the 50 Hz noise is from the lab environment, as similarly observed in trapped ion experiments<sup>30,31</sup>. Mitigating the ambient noise entirely likely requires a combination of additional magnetic shielding, physically relocating the sources of 50 Hz noise, and active stabilisation of the magnetic field.

The developed high-purity (111)-oriented growth in isotopically engineered diamond might create new opportunities for quantum simulations of many-body physics, where coupled spin ensembles with preferential alignment are beneficial<sup>28</sup>. Finally, the record coherence times demonstrated might lead to further improved spin-qubit control for quantum networks based on colour centre qubits<sup>3,79</sup>.

## Methods

### Diamond growth setup

All diamond epilayers were grown using a MPCVD system (SDS6300, Seki Diamond Systems, Cornes Technologies Ltd.) equipped with a 2.45 GHz magnetron and a turbomolecular pump. Diamond substrates were mounted on a cylindrical molybdenum holder (12 mm diameter with a central counterbore), ensuring a run-to-run variation of the diamond surface height relative to the holder surface within  $\pm 100 \mu m$ . This mounting geometry was used to maintain a similar plasma-sample distance and sample geometry across growth runs, thereby reducing run-to-run variations in the near-surface plasma chemistry.

The surface temperature was controlled by the microwave input power ( $< 500$  W) and monitored in situ through a viewport using an optical pyrometer DWF-24-40-C (Williamson), or DFP-2000 (Spectrodyne Inc.).

Process gases consisted of hydrogen ( $> 99.999\%$ ), methane with natural  $^{13}C$  abundance ( $^{13}C = 1.1\%$ , purity  $> 99.9995\%$ ), and isotopically enriched methane ( $^{12}C > 99.99\%$ , purity  $> 99.9\%$ ; Cambridge Isotope Laboratories). Gas purifiers PS7-PD05 for  $H_2$  and PS4-MT3 for  $CH_4$  (SAES Pure Gas Inc.) were used to minimize residual impurities, such that the nitrogen concentration in the mixed gas was expected to be a few ppb or lower, based on the manufacturer specifications and the low methane concentration ( $CH_4/H_2 < 0.1\%$ ).

### Isotopically controlled samples

Undoped and isotopically engineered diamond epilayers (samples *Fukuoka*, *Kagawa*, and the calibration sample *LD3*) were grown homoepitaxially on low-dislocation high-pressure high-temperature (HPHT) type-IIa diamond substrates. The HPHT substrates (Technological Institute for Superhard and Novel Carbon Materials) were used to reduce dislocation density and background photoluminescence from substrate NV centers. The concentration of P1 centres (substitutional nitrogen) in the substrates, estimated by electron spin resonance, was approximately 1 ppm.

Hydrogen flow rates were set to 300 sccm for *Fukuoka* and 2000 sccm for *Kagawa* and *LD3*. Methane flow rates were controlled using mass-flow controllers FCST1000MF series (Fujikin Inc.), yielding  $CH_4/H_2$  ratios of approximately 0.03% for *Fukuoka*, 0.08% for *Kagawa*, and 0.01–0.08% for *LD3*. Growth was performed at a total pressure of 120 Torr and a surface temperature of  $915 \pm 25^\circ C$  with microwave power below 500 W. The low  $CH_4/H_2$  ratios used for these samples resulted in relatively slow growth rates of  $< 0.7 \mu m/h$ . These conditions were chosen to maintain a step-flow-dominated growth mode with suppressed secondary nucleation on the (111) surface<sup>80</sup>.

### Intentionally nitrogen-doped samples

Three intentionally nitrogen-doped diamond samples (S1–S3) were grown on Sumicrystal® HPHT type-Ib diamond substrates (Sumitomo Electric).

Growth conditions were fixed at  $\text{CH}_4/\text{H}_2 = 0.05\%$ , a total pressure of 100 Torr, and a microwave power of 420–490 W.

Samples S1 and S2 were grown under identical conditions, each containing two nitrogen-doped layers formed with gas-phase  $\text{N}_2/\text{CH}_4$  ratios of  $1.2 \times 10^5$  and  $5 \times 10^5$  ppm. These conditions yielded nitrogen concentrations of 8 and 47 ppm in S1, and 7 and 43 ppm in S2. Sample S3 was grown with a gas-phase  $\text{N}_2/\text{CH}_4$  ratio of  $1.0 \times 10^6$  ppm, resulting in a nitrogen concentration of 72 ppm.

The measured N concentration follows an approximately linear power-law,  $[\text{N}] \propto (\text{N}_2/\text{CH}_4)^{1.08}$ , similar to the  $\propto (\text{N}_2/\text{CH}_4)^{1.00}$  dependence reported by Teraji et al. for (100) growth<sup>46</sup>, albeit with a reduced prefactor. This near-linear scaling implies an approximately constant incorporation efficiency over the measured range

$$\eta_{\text{lower}} = 7.5_{-2.6}^{+2.6} \times 10^{-5}, \quad (13)$$

where the uncertainties indicate 95% confidence intervals.

### Isotope quantification and calibration

Carbon isotope compositions are conventionally expressed as the  $^{13}\text{C}/^{12}\text{C}$  ratio relative to the Vienna Pee Dee Belemnite (VPDB) standard<sup>81</sup>. Throughout this work, we use the  $^{13}\text{C}$  concentration  $\chi = ^{13}\text{C}/(^{12}\text{C} + ^{13}\text{C})$ ;  $\chi$  is related to the isotope ratio  $R = ^{13}\text{C}/^{12}\text{C}$  via  $\chi = R/(1 + R)$ .

To place  $\chi$  on an absolute scale, a multi-layer calibration structure with different  $^{13}\text{C}$  concentrations (sample LD3) was analysed by SIMS. To avoid possible bias from gas-memory effects during switching between natural-abundance and  $^{12}\text{C}$ -enriched methane, SIMS ratios were not referenced directly to VPDB. Instead, instrumental offsets were removed using  $A = R_{\text{VPDB}}/R_{\text{Ref}}$  where  $R_{\text{VPDB}} = 0.011113 \pm 0.000022$  is the IUPAC-recommended value for VPDB<sup>81</sup>, and  $R_{\text{Ref}}$  is the SIMS isotope ratio of an external natural-abundance type-IIa {100} CVD diamond reference. The  $^{13}\text{C}$  concentration of the calibration sample LD3 layers was then obtained as

$$\chi = \frac{A \cdot R_{\text{LD3}}}{1 + A \cdot R_{\text{LD3}}}. \quad (14)$$

The natural-abundance layers of sample LD3 ( $\chi_1 = 1.0937(86)\%$ ) were compared with the external natural-abundance {100} reference diamond using the isotope delta notation<sup>82</sup>,

$$\delta_{\text{LD3/Ref}} = \left( \frac{R_{\text{LD3}}}{R_{\text{Ref}}} - 1 \right) \times 1000 \approx -5\text{‰}. \quad (15)$$

This small negative offset may arise from gas-memory effects during methane switching, variations in the isotopic composition of the natural-abundance methane supply, SIMS-related systematics, or a combination thereof. Even if the full magnitude of the observed offset were solely attributed to gas-memory-induced mixing and were to affect the  $^{12}\text{C}$ -enriched layers in an analogous way, the measured  $\chi_0$  could be overestimated by at most about 0.06 ppm. This is negligible compared with the  $\pm 3$  ppm measurement uncertainty. We therefore find no evidence for a measurable gas-memory effect and quote  $\chi_0 = 13(3)$  ppm as our best estimate on the VPDB scale.

For completeness, the isotope delta of the {100} reference diamond relative to VPDB is

$$\delta_{\text{Ref/VPDB}} = \left( \frac{R_{\text{Ref}}}{R_{\text{VPDB}}} - 1 \right) \times 1000 = +13.2\text{‰}. \quad (16)$$

### Nanofabrication

Nanofabrication of SILs and striplines was performed on Fukuoka. First, room-temperature pulsed ESR was used to identify spatially isolated NV candidates. SILs were fabricated using a dual-beam Focussed Ion Beam (FIB) setup. High-power laser exposure of the surface during the optical

characterization was used to leave recognizable markings under electron-beam microscopy, to align in the dual-beam FIB setup. Because the surface exhibited recessed pit-like features with a terraced structure, likely formed by crystallographically anisotropic etching and subsequent overgrowth during step-flow growth, a novel method to pattern gold was used. The striplines were patterned using direct laser lithography, aligning on the SILs, with negative resist and liftoff. The gold was evaporated under an angle on a rotating platform to ensure step-coverage. Direct laser lithography and liftoff of an ALD deposited  $\sim 100$  nm  $\text{AlO}_x$  layer was performed to act as an anti-reflective coating, but this layer was damaged during lift-off.

### NV Experimental Setups

Measurements of bulk NV centres on both Kagawa and Fukuoka (Fig. 3a–c, grey data in d) were performed on a first setup, while all other NV spin and optical measurements were performed on a second setup on an NV in a SIL in Fukuoka. Both setups consist of a Montana Cryostation S50 to cool the diamond to 4 K, with a confocal microscopy setup for optical collection (avalanche photon detector) and excitation (AOMs used for pulse shaping). Microwave pulse generation is performed with a ZI HDAWG arbitrary waveform generator and an R&S SGS100A MW source. For bulk NVs square pulses were used, for the SIL NV hermite-shaped pulses were used. After MW amplification a MW switch is used to minimize noise that may flip or dephase the electron spin<sup>20</sup>. The primary difference between the setups is their location in the building, the position of equipment with respect to the cryostat, and the usage of inner-outer DC blocks in the second setup to mitigate ground loops, which may explain the difference in Hahn-echo signal between the setups (Fig. 3d), and the slightly longer  $T_2^*$  in the second setup (Fig. 3g). This implies that the 50 Hz interference originates both from current running through the stripline and from ambient fields, which for example can be caused by power supplies of lab equipment<sup>30</sup>. All data was taken using the QMI Python package<sup>83</sup>.

All spin measurements consisted of a (1) charge-resonance (CR) check<sup>49</sup>, (2) a laser pulse resonant to the  $E_{\pm 1}$  transition to initialize the ground-state spin, (3) a sequence of MW pulses, (4) a laser pulse resonant to the  $E_y$  or  $E_x$  transition to readout the spin state in a single shot. Typical average single-shot readout fidelities<sup>71</sup> in bulk lie around 65% (in both samples), for the SIL NV in Fukuoka this is up to 92.5% (20 nW laser power, 8  $\mu\text{s}$  pulse length). The reported spin-state readouts are corrected for the known SSRO infidelity.

For the CPMG data the phase of the final  $\pi/2$  pulse was alternated between 0 and  $\pi$  and the difference between the two resulting estimated electron-spin-state populations was used to correct for drifts in photon collection efficiency. For presentation purposes, the CPMG data and fits (Fig. 4b and Supplementary Note 6) were rescaled by the fit parameter  $A$ , which ranges from 0.9 to 1.1. The Ramsey data of bulk NVs (Figs. 3a–c) was measured using slow  $\pi/2$  pulses, so that an oscillation only occurs when the nitrogen spin is in the  $m_I = 0$  state. For the SIL NV (Fig. 3g), fast  $\pi/2$  pulses were used, and the data was post-processed with a 100 kHz low-pass filter to eliminate the  $\sim 2$  MHz oscillations due to the  $m_I = \pm 1$  states.

The magnetic field for bulk measurements on Fukuoka (Kagawa) was  $\sim 1.5$  mT ( $\sim 2.5$  mT), and for measurements on the SIL NV in Fukuoka it was  $\sim 3.6$  mT. Magnetic fields were applied with an external permanent magnet and aligned along the surface-normal [111] NV ( $z'$ ) axis. The field strengths are large enough to place the NVs in the strong-field regime in which perpendicular hyperfine couplings of the nuclear spins may be ignored in the Ramsey experiments<sup>55</sup>. The magnetic field required for this regime scales as  $\chi$  because the typical nuclear spin couplings similarly scale as  $\chi$ , and is therefore much lower than for natural abundance.

### Data availability

The data underlying this study are available on the open 4TU data server [84].

## Code availability

The analysis code underlying this study is available on the open 4TU data server<sup>84</sup>.

Received: 15 April 2026; Accepted: 2 September 2026;

Published online: 28 September 2026

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## Acknowledgements

We thank Yuhei Sekiguchi and Hideo Kosaka for performing preliminary low-temperature PLE measurements on diamond samples. We thank Satoshi Koizumi and Philippe Bergonzo for fruitful discussions on diamond growth. We also thank Ryoichi Ishihara for selecting the CVD-related equipment, and Charles de Boer for assisting with its installation in the Kavli Nanolab cleanroom at Delft University of Technology. We thank Breno Perlingeiro Correa and Georgios Delaroudis for their contribution to the nanofabrication on the Fukuoka sample. We thank Peter Schaefer for his assistance in preliminary room-temperature characterisation of the NV samples. We thank Pieter Botma for preparing the spectral diffusion measurements for the NV center from Ref. [13]. We thank Régis Méjard for maintenance support on one of the low-temperature measurement setups. We thank Mohamed Abobeih and Guido van de Stolpe for discussions. We acknowledge funding from the Dutch Research Council (NWO) through the project “QuTech Phase II funding: “Quantum Technology for Computing and Communication” (Project No. 601.QT.001). This project has received funding from the European Union’s Horizon Europe research and innovation program under grant agreement No 101135699. We gratefully acknowledge support from the joint research program “Modular quantum computers” by Fujitsu Limited and Delft University of Technology, co-funded by the Netherlands Enterprise

Agency under project number PPS2007. This work was supported by the Netherlands Organization for Scientific Research (NWO/OCW), as part of Quantum Limits (project number SUMMIT.1.1016). We also acknowledge support from the Japan Science and Technology Agency (JST), Moonshot R&D Program (JPMJMS2062).

### Author contributions

T.Y. conducted the diamond growth experiments, while B.v.O. and K.-N.S. performed the optical and spin measurements. B.d.Z. assisted in preliminary room-temperature characterisation of the NV samples. S.S. measured the nitrogen concentration in the type IIa HPHT substrate, and S.O. and T.O. carried out the electron beam irradiation experiments. R.V. and H.A.-T. fabricated the SIL device. T.Y. and B.v.O. wrote the paper, with input from T.H.T. and K.-N.S.; T.H.T. supervised the overall research.

### Competing interests

The authors declare no competing interests.

### Additional information

**Supplementary information** The online version contains supplementary material available at <https://doi.org/10.1038/s41534-026-01368-y>.

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