

Millisecond-Coherent 2D Nuclear Shells around V_B^- Defects in hBN

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We demonstrate an essentially two-dimensional (2D) driven central-spin regime based on negatively charged boron vacancy (V_B^-) defects in the dense nuclear quadrupolar lattice of hexagonal boron nitride (hBN). At 3 T magnetic field, the ^{11}B bath collapses into classes of planar, equidistant nuclei. Electron-nuclear double resonance spectroscopy and *ab initio* calculations show that the electron spin density is in fact localized in the defective boron nitride layer exclusively, and that this 2D geometry reduces the electron-nuclear spin Hamiltonian to a function of discrete shell coordinates within the nanometer-scale vicinity of the defect. Even the deviations from bulklike quadrupole coupling of ^{11}B reveals an abrupt truncation across the van der Waals gap. V_B^- -hBN is thus established as a platform of discrete 2D nuclear shells, enabling shell-selective coherent control of ^{11}B nuclei with $T_{2n}^* \approx 100\ \mu\text{s}$ and shell-dependent Hahn-echo coherence times T_{2n} spanning 0.33–1.44 ms.

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The exploration of electron-nuclear spin systems lies at the heart of solid-state quantum technology, including quantum sensing, information, and many-body spin physics [1–7]. In 3D platforms such as nitrogen vacancy (NV) centers in diamond, spin defects in SiC, or donors in silicon, the central spin couples to sparse nuclear environments, allowing these nuclei to function as quantum memories and registers with long coherence times [1,2,4,8–10]. Because magnetic bath isotopes occupy a dilute and statistically varying subset of lattice sites, the microscopic register is generally configuration specific. Even for equidistant nuclei, the dipolar hyperfine (HF) contribution varies with the lattice-defined angle θ as $\propto (1 - 3\cos^2\theta)/r^3$ [11]. This angular variance, combined with a $1/r^3$ estimate of the dipolar contribution, gives a logarithmic sensitivity to the bath size, $\sim \ln(R/a)$, where a and R represent the short-range lattice-related and long-range system-related cutoffs, respectively [12]. In the opposite limit, III–V quantum dots feature a dense environment of ubiquitous quadrupolar nuclei ($I > 1/2$), where HF and nuclear quadrupolar (NQ) interactions dominate the spin response [6,13,14]. Ultimately, the inherent geometry

of the 3D lattice defines a coupling environment where the spectral identity of nuclei is lost within a broad collective background [6,11,13–15].

Layered van der Waals (vdW) crystals such as hexagonal boron nitride (hBN) provide a distinct setting for central-spin physics. The weakly coupled sp^2 -bonded planes of hBN form a bipartite honeycomb lattice composed entirely of spinful quadrupolar nuclei (99.9% ^{14}N , $I = 1$; 80.2% ^{11}B , $I = 3/2$; and 19.8% ^{10}B , $I = 3$). Among its optically addressable defects [16–19], the negatively charged boron vacancy, V_B^- , provides an $S = 1$ central spin within D_{3h} symmetry and an axial zero-field splitting of $D \approx 3.46$ –3.65 GHz, while providing optical spin initialization and readout schemes similar to those of the NV center in diamond [16,20–22].

For $\mathbf{B} \parallel c$, symmetry-equivalent nuclei of a given isotope within each crystallographic shell share the same anisotropic HF projection and NQ shift, making each shell transition collectively addressable at a single resonance frequency.

The vdW gap is expected to suppress the out-of-plane extension of the defect state, so that the dominant defect-induced anisotropic HF interaction inherits the quasi-two-dimensional geometry of the defect-containing layer. In contrast to 3D systems, the dipolar HF sum in this geometry formally converges as $\sim 1/a$, because the $1/r^3$ decay of the interaction outpaces the linear growth in the number of nuclei within a radial shell of fixed thickness. Together, these constraints collapse the bath into a shell-resolved structure, effectively suppressing the accumulation of a stochastic background.

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According to Ref. [23] the HF splitting of remote ^{14}N nuclei rapidly converges with distance from the defect center. Similarly the dense, i.e., ^{11}B - and ^{10}B -spin-complete, B sublattice of $V_{\text{B}}^-:\text{hBN}$ defines an intermediate central-spin regime between configuration-specific individual nuclear registers in diamond and SiC and unresolved mesoscopic nuclear ensembles in III–V quantum dots.

Previous studies of the V_{B}^- center have mainly focused on the defect core, namely the three nearest-neighbor nitrogen nuclei with strong HF couplings, for polarization and coherent control [24–26]. More distant ^{14}N nuclei have been probed through their impact on the V_{B}^- spin coherence [23], and coherent control of a single impurity ^{13}C nuclear spin was recently demonstrated [19], highlighting the potential of hBN for nuclear-spin manipulation. The nuclear environment of V_{B}^- has hitherto been mainly viewed in the context of electron-spin decoherence [27–30].

In this combined experimental and theoretical work, in contrast, we probe its extended nuclear degrees of freedom as a structured and potentially coherent spin system. We performed pulsed electron spin resonance (ESR) and electron-nuclear double resonance (ENDOR) high-frequency spectroscopy (~ 94 GHz, 3 T) at 25 K using a Bruker Elexsys E680 spectrometer and joint Davies and Mims protocols (insets in Fig. 2 and Supplemental Material [31]) [41,42]. The measured parameters for HF and NQ splittings are compared with density-functional theory (DFT) values calculated using norm-conserving *multiprojector* gauge-including pseudopotentials [31] as implemented in the QUANTUM ESPRESSO package [33,34]. The V_{B}^- defect is modeled within 799-atom supercells [see Figs. 1(a) and 1(b)] using a shifted $(2 \times 2 \times 2)$ k -point sampling and a plane-wave basis set with 600 eV energy cutoff. The Heyd-Scuseria-Ernzerhof (HSE) hybrid functional [35] is used to describe exchange and correlation effects of the many-particle system, while vdW interactions are taken into account via Grimme D3 with Becke-Johnson (BJ) damping [36].

V_{B}^- defects were generated in $\sim 150\text{-}\mu\text{m}$ -thick hBN single crystals by 2 MeV electron irradiation ($6 \times 10^{18} \text{ cm}^{-2}$), corresponding to a V_{B}^- concentration of $\sim 6 \times 10^{17} \text{ cm}^{-3}$ [43,44]. The 3 T magnetic field isolates nitrogen and boron sublattices via nuclear Zeeman interactions, enabling selective focus on ^{11}B ($I = 3/2$) by avoiding ^{10}B spectral multiplicity. Under 532 nm excitation, the ESR spectrum of the hBN sample identifies the $S = 1$ V_{B}^- defect. This state is characterized by the zero-field splitting parameter $D \approx 3.57$ GHz and an optically induced polarization of the $m_s = 0$ sublevel [16,20], arising from spin-dependent recombination pathway, as shown schematically in the inset of Fig. 1(c). The resulting population imbalance is reflected by the inversion of the ESR line intensities.

The ^{11}B ENDOR spectra measured on the V_{B}^- $m_s = 0 \leftrightarrow +1$ ESR transition with $\mathbf{B} \parallel c$ are shown in Fig. 2. The spectra exhibit multiple resolved triplet structures related to the Larmor frequency $\nu_L = \gamma_n B / 2\pi \approx 43.98$ MHz, where γ_n

is the nuclear gyromagnetic ratio for ^{11}B . Distinct intensity patterns between the Mims and Davies spectra are observed, with some triplets suppressed in the latter, likely reflecting electron-nuclear spin dynamics in the V_{B}^- center.

Spin-Hamiltonian parameters are determined from

$$\hat{H} = \frac{\gamma_e}{2\pi} B S_z + D \left(S_z^2 - \frac{S(S+1)}{3} \right) + A_{\parallel} S_z I_z - \frac{\gamma_n}{2\pi} B I_z + P \left(I_z^2 - \frac{I(I+1)}{3} \right), \quad (1)$$

where $S = 1$ and $I = 3/2$ are the V_{B}^- electron and ^{11}B nuclear spins, γ_e is the V_{B}^- gyromagnetic ratio, and S_z and I_z

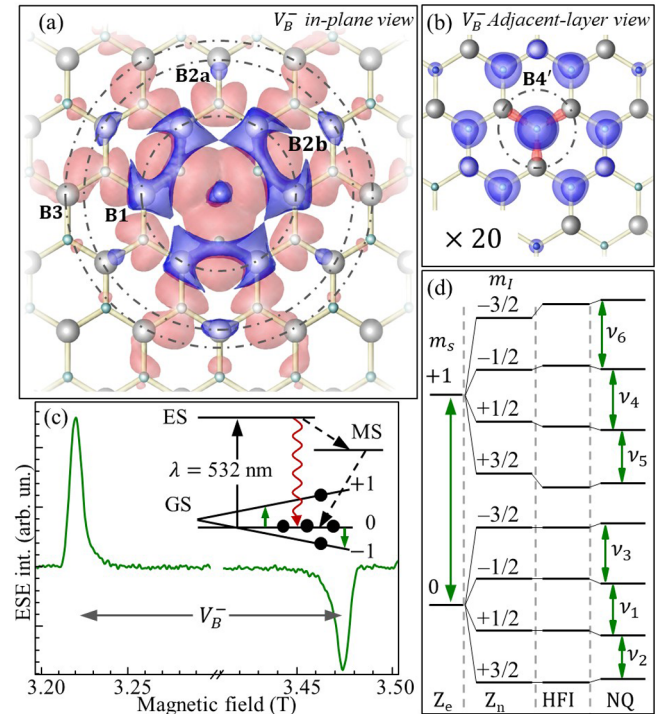


FIG. 1. (a) hBN lattice containing the V_{B}^- defect (center) with the nuclear spin environment: neighboring boron atoms labeled B1, B2, and B3 are organized in rings of six equidistant B nuclei (indicated by dash-dotted circles) whereby B2 splits into two inequivalent sets (B2a, B2b) experiencing different electronic magnetization density $m(\vec{r}) = n^\uparrow(\vec{r}) - n^\downarrow(\vec{r})$ (positive, red; negative, blue). (b) In total six B atoms located in the two adjacent hBN layers are indicated as B4'; magnetization density enlarged by a factor of 20. (c) ESR spectrum of the V_{B}^- center ($\mathbf{B} \parallel c$) under 532 nm excitation; horizontal arrows indicate V_{B}^- triplet transitions. Inset: optical spin polarization into the $m_s = 0$ ground state (GS) via spin-dependent recombination (dashed arrows) through excited (ES) and metastable (MS) states. Purple arrows denote radiative transitions. (d) Energy levels of an $S = 1$ spin coupled to a ^{11}B nucleus, including electron and nuclear Zeeman (Z_e, Z_n), HF, and NQ interactions. The green arrow marks the $m_s = 0 \leftrightarrow +1$ ESR transition at 3221.1 mT. Short green arrows indicate allowed ENDOR transitions ν_{1-6} .

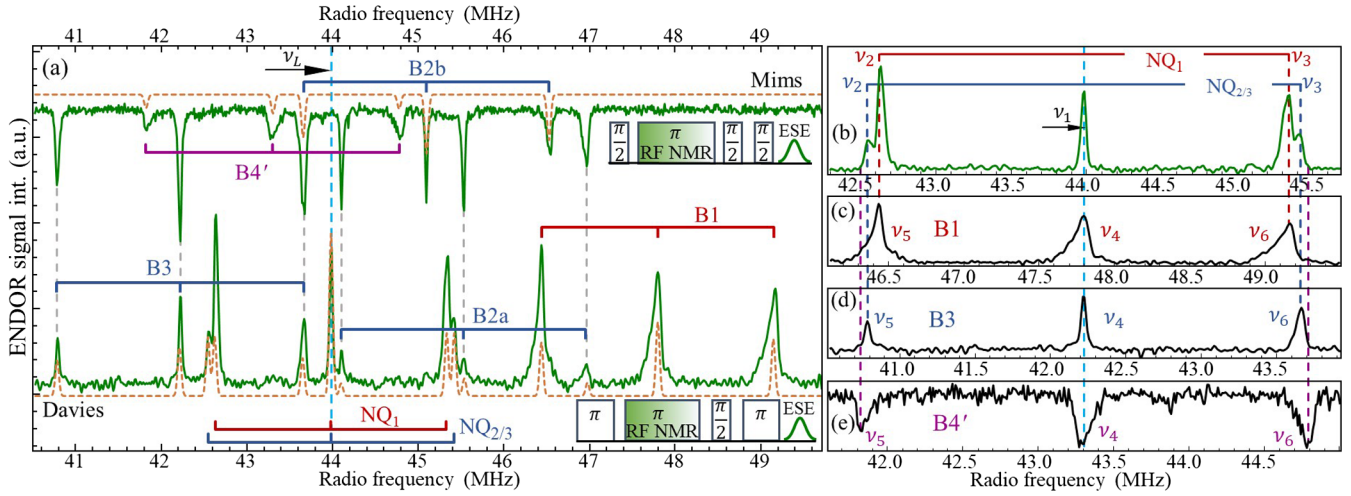


FIG. 2. (a) ^{11}B Mims (top) and Davies (bottom) ENDOR spectra, arising from rf-induced coherence and population changes via the electron spin, respectively; insets show the corresponding pulse sequences. Simulations (dashed orange) are obtained via Eq. (1) and Table I. The Larmor frequency, $\nu_L = 43.98$ MHz, is indicated by the arrow. NQ interaction-induced triplets at ν_L are denoted NQ_1 and $\text{NQ}_{2/3}$, while HF-shifted triplets from different ^{11}B coordination shells are labeled B1–B4' (Davies and Mims ENDOR frequencies indicated by vertical dashed lines). (b) Enlarged view of the $\text{NQ}_1/\text{NQ}_{2/3}$ triplets. (c)–(e) Enlarged views of the B1, B3, and B4' triplets, with transitions labeled according to Fig. 1(c). Blue and red dashed lines highlight the equality of the quadrupole splittings of the B1 and B3 shells with those of NQ_1 and $\text{NQ}_{2/3}$, respectively. The largest NQ splitting belongs to the B4' shell (dashed purple lines); ν_L is marked by the dashed blue line. All spectra in (b)–(e) are shown on the same frequency scale after subtraction of unrelated signals.

are the spin operators along the external magnetic field $\mathbf{B}$, $A_{\parallel}$ is the HF coupling, and $P = 3C_q/4I(2I - 1)$, with $C_q = eQ_n V_{zz}/h$ the NQ coupling constant arising from the interaction of the ^{11}B nuclear quadrupole moment eQ_n with the principal component V_{zz} of the electrical field gradient. For $\mathbf{B} \parallel c$, the principal axes of the zero-field splitting and electrical field gradient tensors coincide with the magnetic field direction, allowing each shell to be described by a single effective nuclear spin $I = 3/2$ characterized by shell-dependent $A_{\parallel}$ and C_q .

Per Eq. (1) and Fig. 1(c), each group of equivalent nuclei exhibits six ENDOR transitions: $\nu_1 = \nu_L$ and $\nu_{2,3} = \nu_L \pm 2P$ in the $m_S = 0$ manifold, and $\nu_4 = \nu_L - A_{\parallel}$ and $\nu_{5,6} = \nu_L - A_{\parallel} \pm 2P$ for $m_S = +1$. The outer triplet splitting, $\Delta\nu = 4P = C_q$, directly gives the ^{11}B nuclear quadrupole coupling constant.

As shown in Fig. 2(b), the triplets NQ_1 and $\text{NQ}_{2/3}$ provide $C_q = 2.79$ and 2.87 MHz, respectively. These values deviate by less than 6% from the intrinsic ^{11}B quadrupole coupling in pristine hBN [37–39], indicating a weak defect-induced perturbation. The triplets B1–B4', shifted from ν_L by shell-specific HF interactions, are shown enlarged in Figs. 2(c)–2(e). The absolute value and sign of $A_{\parallel}$ are determined from the frequency shift between the centers of the $m_S = 0$ and $m_S = +1$ triplets ($\nu_4 - \nu_1$). Specifically, the B1 triplet [Fig. 2(c)] is shifted to a higher frequency, corresponding to $A_{\parallel} \approx -3.8$ MHz, while maintaining a C_q identical to NQ_1 . The B3 [Fig. 2(d)] is shifted to a lower frequency ($A_{\parallel} \approx +1.75$ MHz), matching

the C_q of $\text{NQ}_{2/3}$. The B4' shell [Fig. 2(e)], resolved only in Mims ENDOR, exhibits the largest $C_q \approx 2.96$ MHz and the smallest $|A_{\parallel}| \approx 0.6$ MHz. The B2a and B2b triplets [Fig. 2(a)] exhibit a shared $C_q \approx 2.87$ MHz, with $A_{\parallel} \approx -1.5$ MHz (B2a) and -1.1 MHz (B2b).

The extracted parameters serve as the basis for the ENDOR spectra simulation in Fig. 2(a); the excellent agreement with DFT (Table I) is used to assign the experimental fingerprints to specific nuclear shells. Almost all B-related HF splittings are predominantly given by the isotropic Fermi-contact term. The B1 shell provides an exception. Here, the HF interaction is due to a p -like in-plane oriented spin density and has mainly anisotropic

TABLE I. DFT-calculated (HSE with D3-BJ vdW correction) and experimentally extracted hyperfine ($A_{\parallel}$) and quadrupole (C_q) parameters (in MHz) for ^{11}B atoms in coordination shells B1–B3 (central layer) and B4' (neighboring layers). N_B denotes the number of equivalent boron nuclei in each symmetry group. Experimental bulk C_q values are taken from Refs. [37–39].

Group	N_{B}	DFT (HSE with D3-BJ)		Experiment	
		$A_{\parallel}$ (MHz)	C_q (MHz), η	$A_{\parallel}$ (MHz)	C_q (MHz)
B1	6	−3.89	2.77, 0.275	-3.82 ± 0.05	2.79 ± 0.05
B2a	3	−1.96	2.89, 0.092	-1.55 ± 0.03	2.87 ± 0.03
B2b	3	−1.37	2.92, 0.116	-1.11 ± 0.03	2.87 ± 0.03
B3	6	+2.03	2.92, 0.089	$+1.76 \pm 0.03$	2.87 ± 0.03
B4′	6	+0.67	2.94, 0.036	$+0.65 \pm 0.05$	2.96 ± 0.05
Bulk			2.94, 0.000		2.96 ± 0.04

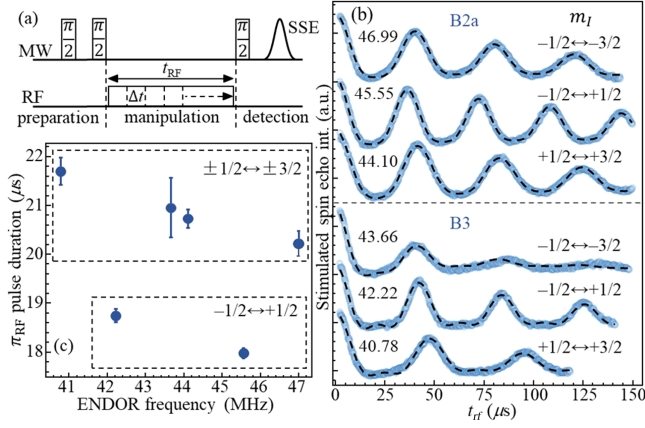


FIG. 3. (a) SSE sequence for nuclear Rabi readout, with a resonant rf pulse of variable duration t_{rf} driving nuclear transitions. (b) Rabi oscillations (transition frequencies in MHz) for selected B2a and B3 ENDOR transitions. Black solid lines are fits to the model. (c) Optimal π -pulse duration versus ENDOR frequency. Data for $\pm 3/2 \leftrightarrow \pm 1/2$ and $+1/2 \leftrightarrow -1/2$ transitions are highlighted by dashed boxes.

character. Despite the 2D confinement of the spin density, the orbital contribution [40] is calculated to remain below the 0.1 MHz limit, indicating orbital quenching even in this quasi-2D regime. Whereas the HF splittings provide a complicated dependence on the distance from the symmetry center, thereby varying nonmonotonously both in value and sign, the quadrupole interaction C_q recovers rapidly toward the bulk value. Remarkably, for the B4' shell located in the atomic layers adjacent to the vacancy, the quadrupole coupling reaches 2.96 MHz. This value falls within the 2.92–3.00 MHz range reported for pristine hBN [37–39], signaling nearly unperturbed lattice conditions. Such recovery across the interlayer space does not only reflect the restriction of the spin density to the defective hBN layer, but also suggests a vdW screening mechanism that effectively confines the defect's electrostatic perturbation. Apart from the contribution of the B4' shell, the spectra show no accumulation of a bulklike quadrupolar response that more distant nuclear shells would produce; instead, the response is dominated by two resolved classes, NQ₁ and NQ_{2/3}. Consequently, the spectra do not evolve into a broad bulklike background, but retain a distinct fingerprint of short-range-dominated 2D-shell behavior. This short-range dominance is quantified by the exact geometric $1/r^3$ lattice sum for the ideal 2D B sublattice. The sum shows that the first three in-plane B shells account for 71.6% of the total ideal in-plane electron-nuclear dipolar HF weight [31]. The traceless anisotropic components of the ENDOR-assigned DFT-calculated HF tensors yield normalized B1:B2:B3 weights of 78.1%:11.6%:10.3%, broadly consistent with the geometric N_s/r_s^3 prediction of 75.9%:14.6%:9.5%, where N_s and r_s denote the number of nuclei and the radius of shell s , respectively [31].

Thus, the geometric lattice sum establishes convergence and near-shell dominance for the ideal 2D sublattice, while the tensor comparison provides a shell-resolved microscopic consistency check for the leading resolved near-field contributions around V_B^- in hBN. This detailed microscopic dipolar HF picture complements recent demonstrations of dimension-dependent many-body dynamics in dipolar electron-spin ensembles of V_B^- [45] and NV defects in quasi-2D diamond slabs [46].

We next demonstrate coherent control of ^{11}B nuclear spins by driving ENDOR transitions with rf pulses of variable duration t_{rf} within a stimulated spin echo (SSE) sequence [Fig. 3(a), [31]], following the scheme proposed in Ref. [47]. The SSE amplitude exhibits oscillations, corresponding to rf-driven nuclear Rabi dynamics; representative data for the B2a and B3 groups are shown in Fig. 3(b). To quantify nuclear decoherence, the oscillations are fitted by $I(t_{rf}) = I_0 e^{-t_{rf}/T_{2n}^*} [1 + C_1 \cos(2\pi t_{rf}/T_{rf}) + C_2 \cos(4\pi t_{rf}/T_{rf}) + I_1 t_{rf}]$. The resulting inhomogeneous dephasing times T_{2n}^* cluster near 100 μ s ([31]), providing a lower bound for the nuclear Hahn-echo coherence T_{2n} . The corresponding Rabi periods $T_{rf} = 2\pi/(\gamma_n B_2 \sqrt{I(I+1) - m_I(m_I+1)})$ follow the scaling expected for the $I = 3/2$ system of ^{11}B ; see also Fig. 3(c). Optimal half-period durations are $T_{rf}/2 \approx 21.17 \mu$ s for the $\pm 3/2 \leftrightarrow \pm 1/2$ and 18.45 μ s for the $+1/2 \leftrightarrow -1/2$ transitions. Their ratio of 1.15 is consistent with the theoretically expected $2/\sqrt{3} \approx 1.15$, providing a robust check of Rabi spin dynamics, independent of absolute rf power calibration. The B3 group exhibits a less regular oscillatory pattern compared to B2a. This increased complexity potentially originates (i) from the indirect nature of the spin polarization at these nuclei (as indicated by the inverted sign of the HF splitting) and (ii) from the higher multiplicity of nuclei (6 vs 3) within the B3 shell. Both effects enhance the sensitivity to inhomogeneous broadening or weak internuclear interactions, giving rise to collective spin dynamics.

To isolate intrinsic coherence from inhomogeneous dephasing, we employ a Davies-ENDOR-based sequence and monitor the nuclear spin echo (NSE) [48,49], cf. Fig. 4 and [31]. From the decay of the NSE intensity we extract $T_{2n} = 1.44, 0.93, 0.91, 0.73$, and 0.33 ms for B1, B2a, B2b, B3, and B4', respectively [Fig. 4(c)]. Notably, the six-site B1 shell shows the longest coherence despite having the shortest B-B distances and, hence, the strongest direct intrashell nuclear dipolar interactions. This may reflect the availability of more nuclear flip-flop pathways. Its anisotropic HF coupling can cause dephasing if the electron spin fluctuates in fixed $m_s = +1$. However, its secular component detunes B1 from the surrounding ^{11}B lattice, while $C_q = 2.79$ MHz suppresses diffusion through satellite transitions. Thus, spectral isolation outweighs electron-induced dephasing in B1, defining the inner region of a

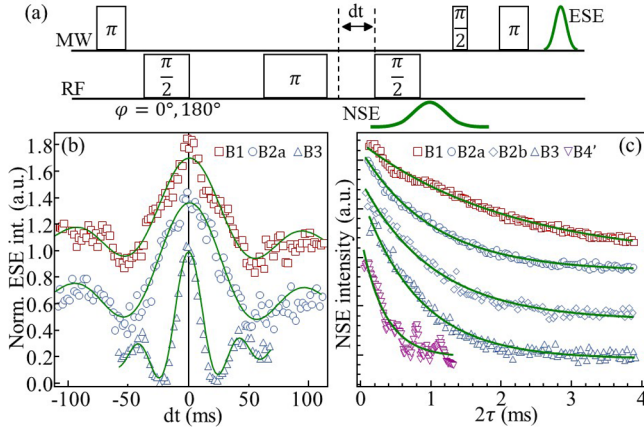


FIG. 4. (a) Davies-type ENDOR sequence for NSE detection. Radio frequency pulses implement a nuclear Hahn echo; phase cycling of the first pulse ($\varphi = 0^\circ, 180^\circ$) isolates the nuclear contribution. (b) Representative NSE signals versus delay dt for the $m_I = -1/2 \leftrightarrow +1/2$ transitions of the shells B1 (47.78 MHz), B3 (42.22 MHz), B4' (43.30 MHz), and for the $m_I = -1/2 \leftrightarrow -3/2$ transition of the shell B2a (46.97 MHz). (c) NSE decay as a function of the total echo time, 2τ . Symbols represent experimental data obtained using the $\pi/2 - \tau - \pi$ sequence. The solid lines are fits to $I(2\tau) = I_0 \exp(-2\tau/T_{2n})$. The extracted T_{2n} values are 1.44 ± 0.05 , 0.93 ± 0.05 , 0.91 ± 0.07 , 0.73 ± 0.05 , and 0.33 ± 0.07 ms for B1, B2a, B2b, B3, and B4', respectively.

quadrupole-assisted frozen core. This resembles strained III–V quantum dots, where quadrupolar shifts are introduced to suppress nuclear-spin diffusion [50]. B2a, B2b, and B3 share the same resolved $C_q = 2.87$ MHz, so no quadrupolar mismatch between their satellite transitions is resolved in $m_S = 0$. At B4', $C_q = 2.96$ MHz matches pristine hBN, removing the quadrupolar mismatch and allowing diffusion into the bulk bath, while $A_{||} = 0.65$ MHz leaves a residual hyperfine barrier. We note that the large Zeeman mismatch between ^{11}B and ^{14}N (44 vs 9.8 MHz) is expected to suppress heteronuclear flip-flops, favoring ^{11}B – ^{11}B spectral diffusion. This is consistent with calculations of the V_B^- coherence in the high-field limit [30].

In summary, we have demonstrated that the hBN lattice and a large axial magnetic field drive the V_B^- defect into a planar central-spin regime where the dense nuclear bath is organized into spectroscopically resolved shells. The observation of bulklike quadrupole coupling for interlayer ^{11}B reveals an abrupt truncation of the defect-induced perturbation across the vdW gap, directly showing how the 2D lattice structure and vdW layering jointly shape the spin environment at the atomic scale. In this 2D geometry, the HF dipolar sum converges, suppressing the accumulation of unresolved couplings and preserving shell resolution [31]. These shells are collectively addressable and retain Hahn-echo coherence for 0.33–1.44 ms, 1 to 2 orders

of magnitude longer than the high-field electron-spin coherence time $T_{2e} \sim 15\text{--}30 \mu\text{s}$ [27–30].

Although shorter than nuclear-spin memories in dilute 3D platforms based on Si, diamond, and SiC [1,2,4,8–10], these coherence times are comparable to those in III–V quantum dots [50,51] and exceed the μs -scale defect-core dynamics previously reported at room temperature [19,24,26,52]. The shell framework can be extended to all isotopes in hBN, opening routes to shell-selective dynamic nuclear polarization, correlated nuclear-cluster states, and collective spin dynamics in 2D materials.

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Data availability—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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