

Probing many-body phenomena with atomically thin nuclear spin layers in diamond

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(Received 18 December 2025; accepted 19 May 2026; published 16 June 2026)

Quantum simulation aims to recreate complex many-body phenomena in controlled environments, offering insights into dynamics that are otherwise difficult to model. Existing platforms, however, are often complex and costly to scale, typically requiring ultrapure vacuum or low temperatures. Here, we introduce a platform based on a thin, strongly interacting ^{13}C nuclear spin layer in diamond that allows controlled exploration of many-body dynamics at room temperature. Nearby nitrogen-vacancy centers enable polarization, readout, and, combined with radio-frequency fields, coherent control of the nuclear spins. We demonstrate strong, tunable interactions among the nuclear spins and use the system to probe discrete time-crystalline order across varying interaction ranges. By combining ease of use with operation at ambient temperatures, our work opens new opportunities for investigating strongly correlated many-body effects.

DOI: [10.1103/6z22-4z36](https://doi.org/10.1103/6z22-4z36)

I. INTRODUCTION

Despite a century of progress, large-scale quantum phenomena remain difficult to study [1]. Quantum simulators, systems that recreate these complex dynamics in controlled environments, allow one to explore such phenomena directly. Recent achievements include simulating cosmological effects [2,3], investigating chemical dynamics [4,5], exploring quantum chromodynamics [6], and creating new phases of matter [7–10]. Most platforms, however, require demanding conditions, such as low temperatures or ultrapure vacuum, raising the following question: Can more accessible systems probe similar dynamics?

Nitrogen-vacancy (N-V) centers in diamond offer optical readout and long coherence times at room temperature [11], and have been used to study many-body phenomena [10,12,13]. Hyperfine coupling to surrounding nuclear spins allows these nuclei to be used as a resource for quantum simulation, provided their interactions are sufficiently strong, i.e., the nuclear spins are densely packed [14]. A dense nuclear spin layer offers the added advantage of forming a self-assembled, spatially ordered spin register that is partially programmable [8], unlike dense N-V center ensembles where disorder and spectral crowding limit controllability. Past efforts to create such dense nuclear spin layers involved surface

termination [14], depositing two-dimensional materials on the diamond [15], and delta doping [16]. None of these systems have yet demonstrated their applicability to probe many-body interactions, due to either rapid depolarization of the nuclear spins or the absence of strong dipolar coupling among them.

In this work, we present a different approach, based on isotopic engineering of diamond through indirect overgrowth [17]. We create a thin ^{13}C nuclear spin layer (≤ 1 nm) next to single N-V centers, embedded in a spin-free ^{12}C environment, where the N-V centers are positioned via low-energy $^{15}\text{N}^+$ implantation during intermediate overgrowth steps. Our method preserves the coherence of both N-V centers and nuclear spins, a major improvement over approaches requiring near-surface N-V centers.

We estimate the distance between N-V centers and ^{13}C layer using a numerical model based on Pauli string truncation, and initialize and read out the nuclear spin layer via the N-V centers. Probing the ^{13}C layer's coherence properties reveals strong, tunable interactions among the nuclear spins, which we harness to demonstrate the system's potential through the generation and study of discrete time-crystalline order [18]. By enabling strong, controllable spin interactions at room temperature, this platform provides a scalable route for exploring strongly correlated quantum phases.

II. SAMPLE FABRICATION AND CHARACTERIZATION

To realize our quantum simulator in a controlled and reproducible manner, we fabricate and characterize two diamond samples. Sample A is implanted after the ^{13}C layer growth, while sample B is implanted before [see Fig. 1(a)]. We begin by indirect overgrowth [17] of a commercial diamond substrate with 96 nm ^{12}C ($I = 0$) in our home-built chemical

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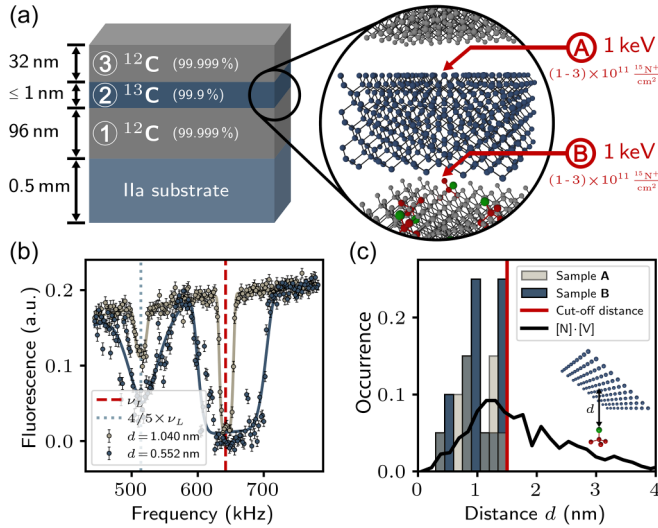


FIG. 1. Sample fabrication and characterization. (a) Structure of our diamond samples with numbered overgrowth steps; layer colors indicate the carbon isotope and its corresponding isotopic purity. The close-up shows an accurate representation of the atomic structure of the ^{13}C layer and the red arrows highlight the implantation steps for samples A and B. (b) AXY spectra of N-V centers with different distances to the nuclear spin layer. The spectra show two dips around the Larmor frequency (red dashed line) and a higher harmonic (dark blue dashed line), with distance-dependent linewidths. (c) Distance distribution for both diamond samples up to the estimation limit, marked by the red line. The black line represents the product of the implantation-induced vacancy density $[V]$ and nitrogen density $[N]$. The inset visualizes the relative angle of the ^{13}C layer to the N-V center's symmetry axis.

vapor deposition (CVD) reactor [19,20]. Sample B is then implanted with 1 keV of $^{15}\text{N}^+$ to create shallow, single N-V centers [21]. Both samples are subsequently overgrown with a thin ^{13}C ($I = 1/2$) layer of ≤ 1 nm thickness after which sample A is implanted. Finally, a 32 nm ^{12}C cap layer is grown to protect the spins from surface noise. The samples are annealed at 1000 °C for 3 h in a ultra-high-vacuum oven to form N-V centers from the implanted nitrogen. A detailed, step-by-step description of the growth process is provided in the Appendix A. Due to the $[100]$ growth, the ^{13}C layer is tilted by 54.7° with respect to the N-V center's symmetry axis.

To analyze the growth process, we probe the N-V center's local ^{13}C environment with the adaptive XY (AXY) dynamical decoupling sequence [22]. By replacing the π pulses of standard dynamical decoupling sequences with a composite pulse, the AXY sequence allows tuning of the interaction strength between the N-V center and surrounding nuclear spins, enabling resolution of distant spins in the presence of close, strongly coupled ones [22]. Each nuclear spin creates a resonance that is shifted from the bare Larmor frequency ν_L by its respective hyperfine coupling [23]. Instead of individual resonances, the large number of ^{13}C nuclei leads to one broad dip around ν_L in the AXY spectra in Fig. 1(b), whose width depends on the distance of the N-V center to the ^{13}C layer. Finite pulse lengths combined with strong A_{zx} and A_{zy} hyperfine interaction lead to an additional harmonic at $4/5 \times \nu_L$ [24].

All experiments are performed at room temperature in a confocal setup, with microwave and rf control applied via a simple copper wire, placed on the diamond's surface [25]. The magnetic field of $B = 600$ G is aligned along the N-V center's symmetry axis.

Using a truncated subspace method in a Pauli string basis, we simulate the measured spectra with a two-dimensional grid of 100 nuclear spins at varying distances from the N-V center [inset, Fig. 1(c)]. With an appropriate truncation [26], we can prioritize Pauli strings of low weight and factor in the different interaction strength magnitudes between the spin species. Our model incorporates any two-spin correlations and all three-spin correlations that involve the N-V center [25]. Equivalent to the experiment, the spectra are fitted with a generalized normal distribution [27], allowing us to correlate the variance with the distance. Because the signal strength scales with the hyperfine interaction, which decreases with increasing distance, it vanishes beyond 1.5 nm (“cutoff distance”) for the chosen AXY settings.

For each sample, we measure the AXY spectrum for 20 different N-V centers and plot the estimated distances as a histogram in Fig. 1(c). Within the measurable range, both samples show a mean distance of 1 nm [25]. For sample A, 50% of the N-V centers fall within this range, compared to 90% for sample B. This is counterintuitive, since sample A is implanted after the ^{13}C overgrowth, so the N-V centers should be closer to the layer than in sample B. CRYSTAL-TRIM [28] simulations of the product of implanted nitrogen and created vacancy densities [black line in Fig. 1(c)] yield 45% below the cutoff distance. The product accurately captures the depth distribution of shallow implanted N-V centers, thus serving as a reasonable reference (see Appendix B).

Because the diamond is exposed to hydrogen plasma before the actual overgrowth begins, these findings suggest that the plasma etches away the outer, possibly damaged diamond layers. This would reduce the distances for sample B and increase them for sample A, implying a ^{13}C layer thinner than the anticipated 1 nm. Variations in the ^{13}C concentration can likewise increase the estimated distances.

III. INITIALIZATION AND CONTROL OF THE NUCLEAR SPIN LAYER

To initialize the ^{13}C layer, we employ the nuclear spin orientation via electron spin locking (NOVEL) method [30], whose pulse sequence is depicted in Fig. 2(a). The Rabi frequency of the spin-lock pulse is adjusted to match the nuclear Larmor frequency, creating an effective flip-flop interaction via the Hartmann-Hahn condition [31]. This allows to pump polarization into the nuclear spin layer through repeated application of the sequence and re-initialization of the N-V center via a laser pulse [29]. Figure 2(a) shows typical measurements for up to $m = 1000$ sequence repetitions. The y axis corresponds to the flip-flop probability $p_{\uparrow_n}(m)$, i.e., the probability to transfer the N-V center's polarization to the nuclear spin layer. The index $\uparrow_n$ denotes into which state the nuclear spins are initialized, which can be switched to $\downarrow_n$ by reversing the sign of the initial $\pi/2$ pulse. Compared to natural abundance diamond [inset, Fig. 2(a)], significantly more polarization steps are required to reach a steady state due to the higher

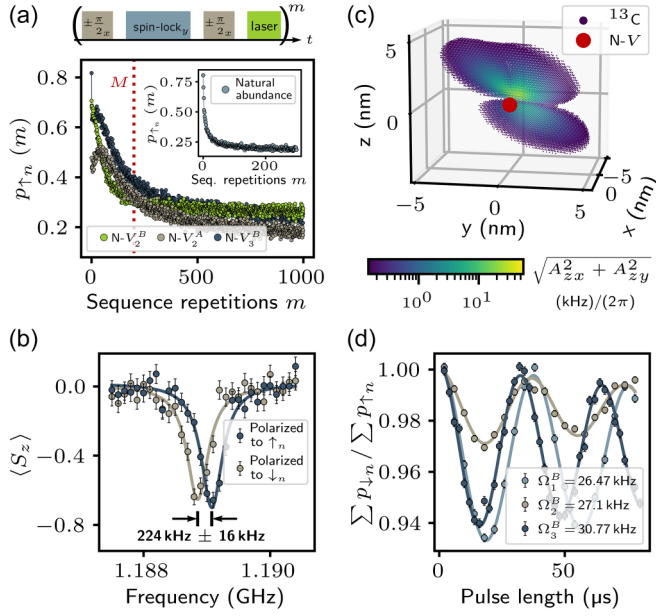


FIG. 2. Initialization and control of the nuclear spin layer. (a) NOVEL pulse sequence used to polarize the nuclear spins with typical measurement results. The y axis corresponds to the N-V center's spin flip-flop probability after m sequence repetitions (x axis) and the inset shows the comparison to a diamond with a natural abundance of ^{13}C . The red vertical line denotes the upper limit M used for the nuclear spin state reconstruction in Eq. (1). (b) ODMR measurement of the Overhauser shift (in nonangular frequency units). The color indicates the initialized state of the nuclear spins. (c) Matching the experimental settings, the image visualizes the addressed nuclear spins during the Overhauser shift measurement, assuming a layer thickness of 1 nm. The color of the nuclear spins corresponds to their perpendicular hyperfine coupling, which is proportional to the flip-flop frequency [29]. (d) Rabi measurements of the ^{13}C layer for different N-V centers, with the pulse sequence. After m NOVEL repetitions to polarize the nuclear spins into $|\uparrow_n\rangle$, an rf pulse is applied, followed by m NOVEL repetitions into $|\downarrow_n\rangle$ for readout. The Rabi frequencies are given in nonangular frequency units. The y axis is proportional to the collective nuclear $\langle I_z \rangle$ expectation value.

number of nuclear spins. Some measurements show a non-exponential decay, which can be attributed to nuclear spins with strong A_{zx} or A_{zy} coupling [25]. Such strongly coupled spins can partially inhibit or even fully block the polarization transfer to the other spins [25,32,33].

Tensor-network simulations show $>90\%$ polarization after 2000 NOVEL repetitions for a planar grid of 25 spins at the estimated mean distance of 1 nm to the N-V center [25]. For more distant nuclear spins, the polarization significantly drops. Consequently, the early data points in Fig. 2(a) are dominated by the nearest spins, and the size of the probed spin environment can be adjusted through the total number of repetitions M .

Polarization of the nuclear spins induces an Overhauser shift [34], measured in an optically detected magnetic resonance (ODMR) experiment via the N-V center [Fig. 2(b)]. Each data point is obtained after 1000 NOVEL repetitions and followed by another 1000 repetitions with an inverted sign of the initial $\pi/2$ pulse to reverse the built-up polarization.

To ensure that setup instabilities do not affect the measured shift, the measurements for $|\uparrow_n\rangle$ and $|\downarrow_n\rangle$ are acquired simultaneously, yielding a shift of $224 \pm 16 \text{ kHz}$. Assuming a 1 nm thick ^{13}C layer at the measured distance of 0.8 nm from the N-V center and neglecting spin diffusion, we estimate an average polarization of $24\% \pm 2\%$ across $\sim 15\,000$ nuclear spins from the observed shift. The estimation only considers spins capable of performing an entire flip-flop within the 1000 NOVEL repetitions, i.e., within $\sim 7 \text{ ms}$ (see Fig. 2(c)). For 15 000 spins, the realistic upper bound for 1000 perfect flip-flops is 6.6% ($= 1000/15\,000$), which is much smaller than the polarization estimated from the Overhauser measurement. This discrepancy is likely caused by nearby spins with strong A_{zz} couplings that enhance the measured shift.

With efficient initialization of the nuclear spins in place, we now focus on their control and readout. All experiments begin by initializing the nuclear spins into $|\uparrow_n\rangle$. Next, the actual experiment is performed, e.g., driving nuclear Rabi oscillations with rf pulses, followed by pumping the spins into $|\downarrow_n\rangle$. For every time step in the Rabi measurements in Fig. 2(d), we obtain two NOVEL curves, equivalent to those in Fig. 2(a). The mean nuclear $\langle I_z \rangle$ expectation value can then be determined from the ratio of summed data points [25],

$$\langle I_z \rangle \propto \sum_m p_{\downarrow n}(m) / \sum_m p_{\uparrow n}(m). \quad (1)$$

IV. LAYER DYNAMICS AND TUNABLE INTERACTIONS

To assess the spin properties of the ^{13}C layer, we first measure the nuclear depolarization [Fig. 3(a)]. During the measurement, the N-V center is periodically reset via a laser pulse to account for its limited T_1 and maintain the initial $|m_s = 0\rangle$ state. The observed depolarization time of $22 \pm 3 \text{ ms}$ is significantly shorter than the expected T_1 at room temperature [35] and attributed to spin diffusion. As polarization spreads and the polarization gradient decreases, diffusion becomes progressively slower, possibly explaining the persistent z magnetization. The residual magnetization may also result from normalization errors, e.g., detuning from the Hartmann-Hahn resonance [25]. Cycling through the N-V center's spin states during its reset may also cause additional depolarization.

Ramsey measurements reveal nuclear spin dephasing times T_2^* in the low hundreds of microseconds for both samples [25], as visualized in Fig. 3(b). The observed variation in T_2^* and line shapes likely arises from inhomogeneities of the ^{13}C concentration. For comparison, nearly 100% ^{13}C -enriched diamond powder yields $T_2^* = 60 \pm 5 \mu\text{s}$ at 7.05 T in an NMR measurement [25]. Apart from inhomogeneities, the longer T_2^* of our ^{13}C layer can be attributed to its thickness of only a few atomic layers, which limits nuclear interactions.

Measuring the nuclear coherence time via a Hahn echo [36] leads to only minor improvements over T_2^* . As seen in Fig. 3(c), oscillations arising from dipolar interactions among nuclear spins persist, since spin echo sequences like Hahn echo are designed to decouple on-site disorder [37], e.g., detuning. Assuming dephasing is dominated by dipolar interactions, a Hahn echo is not expected to extend the coherence time.

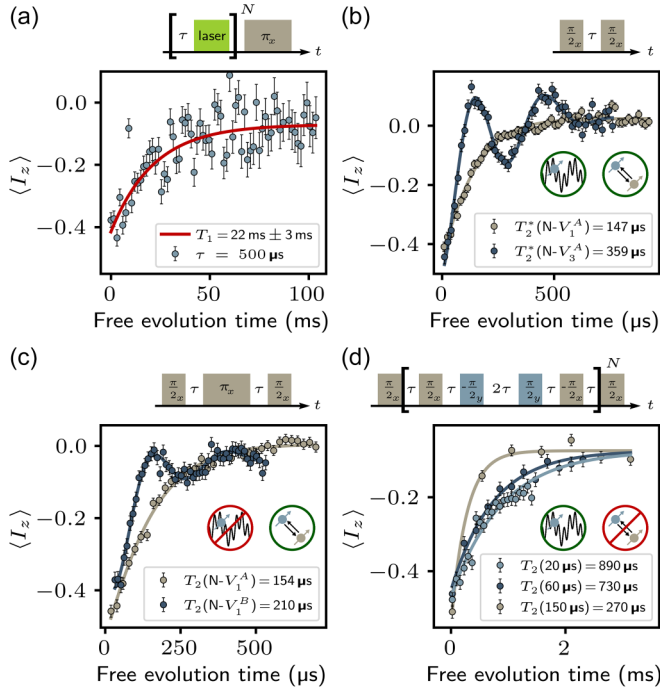


FIG. 3. Spin properties of the ^{13}C layer. (a) Depolarization lifetime measurement in sample B, showing a significantly reduced T_1 . The free evolution time (x axis) is varied by repeating a free evolution period τ , followed by a laser pulse that resets the N-V center's spin state to $m_s = 0$, for a total of N repetitions. (b) Ramsey measurements with corresponding pulse sequence. During the experiment, the nuclear spins are subject to on-site disorder (first pictogram from left) and nuclear dipole-dipole interactions (second pictogram). The notation N-V_k^S indicates the sample S and the N-V center k used in the measurement. (c) Designed to suppress on-site disorder, the Hahn echo measurements show minimal improvements and exhibit oscillations attributed to nuclear dipole-dipole interactions. (d) Homonuclear decoupling through the WAHUA sequence efficiently decouples the nuclear spins from each other. The different colors correspond to different pulse spacings τ .

In contrast, homonuclear decoupling sequences like WAHUA [37,38], which decouple dipolar interactions, significantly increase the coherence time, as illustrated by Fig. 3(d). Effective decoupling requires pulse spacings τ much shorter than T_2^* . If τ approaches T_2^* , the decoupling fails. The WAHUA sequence not only confirms strong interactions among the nuclear spins but also allows to tune their interaction strength by varying τ . At short τ , our measurements are limited by rf heating due to the increased duty cycle.

V. CREATION OF DISCRETE TIME-CRYSTALLINE ORDER

To demonstrate the ^{13}C layer's applicability to explore many-body interactions, we use it to probe time-crystalline order. A time crystal is a nonequilibrium phase of matter that spontaneously breaks time-translation symmetry [39,40]. It can occur in many quantum systems, most notably periodically driven, so-called Floquet systems [8–10], where it manifests as a discrete time crystal (DTC), exhibiting oscil-

lations at multiples of the driving period that persist under perturbations [18].

To create a DTC with our ^{13}C layer, we apply the pulse sequence shown in Fig. 4(c). Each Floquet cycle N ($\equiv$ one sequence repetition) comprises a rotation pulse $R(\theta)$, with angle θ placed between two free evolution times $U(\tau)$ of length τ . After each $U(\tau)$, a laser pulse resets the N-V center to $m_s = 0$. Measurements for sample B are shown in Fig. 4(a), choosing $\theta = 1.03\pi$ for varying τ . In a noninteracting system, the 3% over-rotation creates a beating that we observe for $\tau = 0 \mu\text{s}$, accompanied by a strong decay. Figure 4(b) shows the corresponding power spectral density of the baseline-corrected measurement data. The baseline itself is determined from the fits shown as solid lines in Fig. 4(a). Increasing τ allows the nuclear spins to interact, forming the time-crystalline phase that stabilizes the oscillation with the expected $2T$ -periodic response, where T is the Floquet period.

By calculating the crystalline fraction $\mathcal{C} = |S(\nu = 1/2)|^2 / \sum_\nu |S(\nu)|^2$ [10], the ratio of the $\nu = 1/2$ amplitude (intensity of the $2T$ -periodic response) to the total spectral power, we can visualize the phase transition in Fig. 4(d). In contrast to experiments utilizing layers of coupled N-V centers [10,12,13], our system allows us to bias the experiment toward different spatial regions and coupling classes of the layer during postprocessing by tuning M [the number of NOVEL repetitions; see Eq. (1)]. As discussed in Sec. III, for small M the dynamics are dominated by the nearest nuclear spins to the N-V center, whereas more distant spins become dominant at larger M . By phenomenologically approximating the observed phase transition with a single exponential (with $R^2 > 95\%$ for all curves), we can extract the $1/e$ time τ_0 for different values of M , to visualize the differences in the observed dynamics in Fig. 4(e). The second x axis of Fig. 4(e) shows the estimated number of nuclear spins read out, assuming a layer thickness of 1 nm and restricting the interaction range to spins capable of completing a full flip-flop within M NOVEL repetitions. For small M , i.e., when the signal is dominated by the nearest nuclear spins, we observe a significantly longer interaction time required for the system to exhibit stable DTC order ($\tau > 3\tau_0$) compared to larger M . This behavior is likely caused by the reduced nuclear-nuclear coupling strength of the nearest spins, which are located at the ^{12}C - ^{13}C interface. In contrast, more distant spins, situated deeper within the ^{13}C layer, experience stronger average coupling and can interact with a larger number of neighboring nuclear spins, leading to a much shorter τ_0 at large values of M . This effect is also evident in Fig. 4(f) at an interaction time of $\tau = 0 \mu\text{s}$, where no DTC order has yet emerged. For $M = 300$, the dynamics differ significantly from those observed at $M = 500$, where variations in coupling strength give rise to additional beatings caused by induced over-rotation ($\theta = 1.03\pi$). At large M , this also results in a slight increase in τ_0 (within the margin of error), which may indicate the presence of inhomogeneities or patchiness in the ^{13}C layer.

Owing to the similarity to prior experiments with dense N-V center ensembles [10], specifically the long-range interaction among the spins, our time-crystalline order is likely stabilized by slow, critical dynamics [41] rather than many-body localization [8]. Because the decay rate of the DTC order

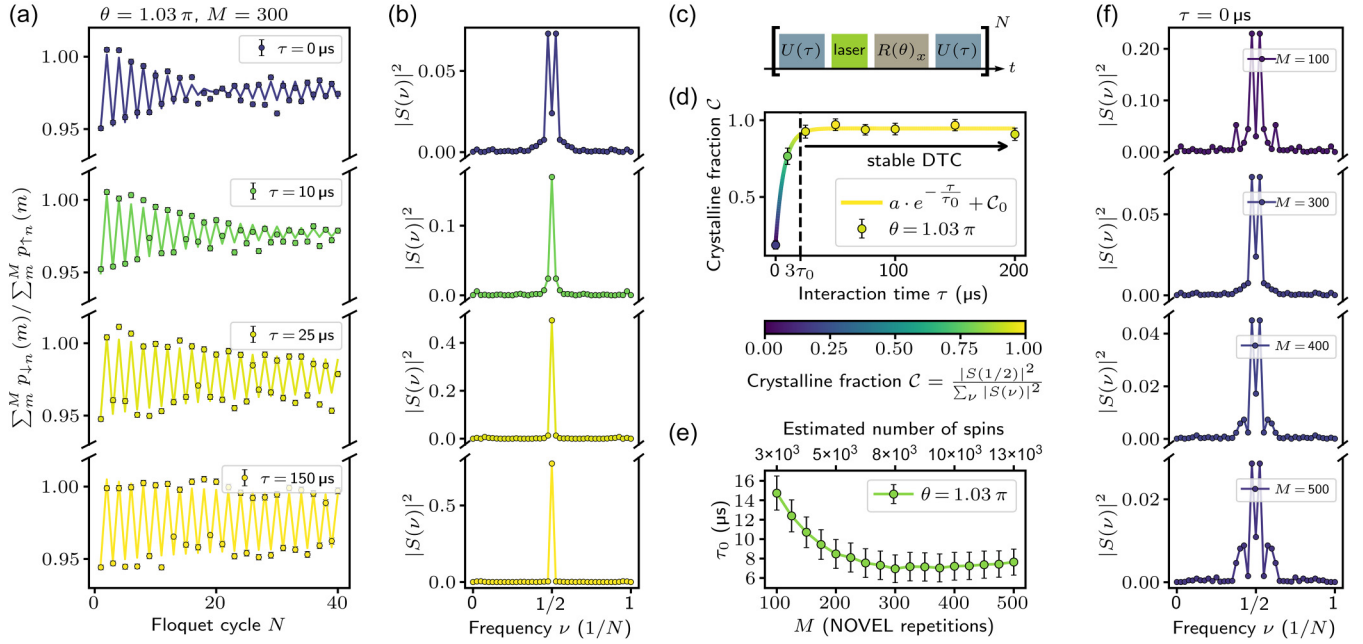


FIG. 4. Exploring discrete time-crystalline order with the nuclear spin layer. (a) Representative DTC measurements for different interaction times τ . For $\tau = 0 \mu\text{s}$, the signal shows a beating caused by the applied over-rotation with rotation angle $\theta = 1.03 \pi$, which vanishes for increasing τ . The x axis corresponds to the Floquet cycles N and the solid lines show the fit result. We use $a \times \cos(\pi p \times N \tau) \times \exp(-(N \tau / T)^\beta) + c$ as the fit function, with a , p , T , β , and c as free parameters. The y axis shows the summed, normalized NOVEL signal ($\propto \langle I_z \rangle$) [see Eq. (1)]. (b) Power spectral density of the measurement data for different interaction times τ . Each row corresponds to the data presented in panel (a). (c) Pulse sequence of the DTC experiment. The blue blocks correspond to a free evolution time for duration τ and the gray block denotes an rf rotation pulse of angle θ . The N-V center is re-initialized into $m_s = 0$ through a laser pulse in each repetition N . (d) Calculating the crystalline fraction C for different interaction times τ allows to visualize the transition to the DTC phase. The measurement data are fitted with a single exponential characterized by a $1/e$ time τ_0 . (e) Extracted values of τ_0 for increasing number of NOVEL repetitions M . The second x axis shows the estimated number of nuclear spins capable of performing a full flip-flop within M NOVEL repetitions. For low M , where spins near the ^{12}C - ^{13}C interface dominate the signal, a stable DTC phase is reached more slowly than for larger M , likely due to reduced average spin-spin coupling in the more dilute interface region. (f) Power spectral density for $\tau = 0 \mu\text{s}$ at different values of M . Varying M probes different dynamics within the layer, reflected in additional beatings arising from the induced over-rotation.

does not increase with interaction time [25], prethermalization [42,43] can also be ruled out as a stabilization mechanism.

Our DTC measurements not only serve as a proof-of-concept demonstration for probing many-body interactions in a large interacting nuclear spin layer, but also highlight the modular readout capabilities of our hybrid N-V- ^{13}C system, enabling the investigation of different dynamics and settings within a single measurement.

VI. CONCLUSION

In conclusion, we present a room-temperature platform to probe many-body phenomena based on an atomically thin nuclear spin layer, along with its fabrication, characterization, and operation. The control could be extended by combining our system with scanning probe techniques, such as magnetic tips [44], to induce strong field gradients that locally modify the layer's Hamiltonian. With an estimated layer thickness below 1 nm, regions with two-dimensional characteristics are within reach and may already contribute to the observations. Such low-dimensional, driven systems are expected to change the relaxation dynamics [41,45], facilitating the investigation of novel phenomena. For example, in a low, tilted magnetic field or in a [111] diamond, all nuclear spins are

oriented perpendicular to the two-dimensional layer, enabling the generation of spin squeezing [13]. Apart from varying the N-V center's sensing volume, changing the ^{13}C concentration also allows to control the disorder, a key parameter when studying time-crystalline order [18,42]. Dynamical decoupling schemes like the showcased AXXY sequence [46] or the dynamically decoupled radio-frequency (DDrf) sequence [47] permit selective control ^{13}C nuclear spins via the N-V center, rendering the system partially programmable.

Given the simplicity and accessibility of our system, our work provides a convenient and scalable platform to study many-body quantum phenomena at room temperature.

ACKNOWLEDGMENTS

We thank Ashok Ajoy, Martin Korzeczek, Michael Olney-Fraser, Priyadarshini Balasubramanian, and Thomas Reisser for helpful discussions. We thank Matthias Poselt and the Helmholtz-Zentrum Dresden-Rossendorf for providing the CRYSTAL-TRIM software. J.P. acknowledges support from the QuantERA II Programme (Mf-QDS) that has received funding from the European Union's Horizon 2020 Research and Innovation Program under Grant Agreement No. 101017733, from Grants No. TED2021-

130578BI00 and No. PID2021-124965NB-C21 funded by MICIU/AEI/10.13039/501100011033 and by the European Union NextGenerationEU/PRTR and from Quantum Flagship project ASPECTS, Grant Agreement No. 101080167. This work was funded by the German Federal Ministry of Research (BMBF) by future cluster QSENS (Grant No. 03ZK110AB) and projects DE-Brill (Project No. 13N16207), SPINNING (Project No. 13N16215), PhoQuant (Project No. 13N16110), DIAQNOS (Project No. 13N16463), quNV2.0 (Project No. 13N16707), QR.N (Project No. 16KIS2208), and EXTRASENS (Project No. 13N16935), DLR via project QUASIMODO (Project No. 50WM2170), Deutsche Forschungsgemeinschaft (DFG) via Projects No. 386028944, No. 387073854, No. 445243414, No. 491245864, No. 499424854, No. 532771161, No. 546850640, and joint DFG/JST ASPIRE Program via Project No. 554644981, European Union's Horizon Europe Program via projects QuMicro (Project No. 101046911), SPINUS (Project No. 101135699), C-QuENS (Project No. 101135359), QCIRCLE (Project No. 101059999), and FLORIN (Project No. 101086142), European Research Council (ERC) via Synergy grant HyperQ (Grant No. 856432), and Carl-Zeiss-Stiftung.

P.J.V., M.B.P., J.P., and F.J. conceived the experiments. P.J.V. conducted and analyzed the measurements of the ^{13}C layer samples and performed the simulations for the time-crystalline order. C.F., C.O., and J.L. fabricated the samples. R.B. performed the NMR measurements and C.F. measured the depths of the 5 keV implanted N-V centers. The CRYSTAL-TRIM simulations were carried out by J.F. M.K. developed the numerical method and model for the distance estimation and produced numerical data that P.J.V. used to determine the distance from experimental data. A.V. and J.P. performed the tensor-network simulations for the polarization dynamics. J.P., M.B.P., and F.J. supervised the work. All authors discussed the results and contributed to the manuscript.

C.F., C.O., J.L., and F.J. are co-founders, C.F. is CTO, C.O. is CFO and COO, and J.L. is CEO of Diatope GmbH. P.J.V. was employed part-time by Diatope GmbH during parts of the project.

DATA AVAILABILITY

All data presented in this study are available from the corresponding authors on reasonable request.

APPENDIX A: OVERGROWTH SETTINGS

In-between all fabrication steps that require the diamonds to leave or re-enter the CVD reactor, the samples are cleaned by a 1:1:1 mixture of nitric (65%), sulfuric (98%), and perchloric (72%) acid for 20 min at 200 °C in a microwave digestion system (ETHOS.lab). After the acid cleaning, they are rinsed with deionized water.

The growth process begins by heating the reactor's sample plate to 700 °C with a graphite substrate heater. We subject the diamonds to a hydrogen plasma for 5 min to ensure the plasma and temperature are in equilibrium before we start the overgrowth. The temperature is monitored by an infrared pyrometer (Optris, model CTlaser 2 MH, wavelength 1.6 μm). During the overgrowth, we reach a tem-

TABLE I. Growth parameters for the different overgrowth steps. The columns show the current step of the overgrowth process and the rows show the corresponding growth parameters.

Parameter/step	1	2	3
Methane gas	$^{12}\text{CH}_4$	$^{13}\text{CH}_4$	$^{12}\text{CH}_4$
Purity	99.999%	99.9%	99.999%
$\text{CH}_4:\text{H}_2$	0.083%	0.025%	0.083%
Flow rate H_2		600 sccm	
Pressure		22.5 mbar	
Microwave power		1.2 kW	
Temperature		900 °C	
Duration	180 min	11 min	60 min
Target thickness	96 nm	1 nm	32 nm

perature of approximately 900 °C. We use isotope-enriched methane from Cambridge Isotope Laboratories with a purity of 99.999% for ^{12}C and 99.9% for ^{13}C . The gases are additionally purified through a palladium filter (Johnson Matthey, Hydrogen Purifier HP-25) and a getter filter for methane (MonoTorr, PS4-MT3-531). Table I summarizes the growth parameters for each overgrowth step. The growth rates are estimated from our earlier work [17]. The diamond surface corresponds to the [100] plane, with sample A exhibiting a miscut angle of 0.3° and sample B 0.1°, as determined by x-ray diffraction (XRD) rocking curve analysis. A comprehensive description of the CVD reactor can be found in Refs. [19,20].

The implantation is performed in a home-built ion implanter [21] that features an IQE12/38 ion source from Specs. To distinguish implanted N-V centers from native ones, we choose a 98% enriched $^{15}\text{N}_2$ gas from Sigma-Aldrich (^{15}N natural abundance: 0.4 %), which is purified by a Wien mass filter during the implantation to ensure that we only implant $^{15}\text{N}^+$. Using an implantation energy of 1 keV, we create several implantation spots with a diameter of 200 μm . We choose three different doses of 1×10^{11} , 2×10^{11} , and $3 \times 10^{11} \text{ }^{15}\text{N}^+/\text{cm}^2$ to create single N-V centers at varying densities after the overgrowth [17].

To finally form the N-V centers from the implanted nitrogen, we place the samples in a home-built ultra-high-vacuum oven at 1000 °C for 3 h with a pressure below 1×10^{-7} mbar during the annealing process.

Before their investigation in a room-temperature confocal microscope, the diamonds are acid cleaned one more time following the cleaning procedure described at the beginning of this Appendix.

APPENDIX B: DISTANCE ESTIMATION

To infer the distance of the N-V centers to the ^{13}C layer from the AXY spectra, we start by numerically simulating the experiment. Equivalent to the experiment, we use 30 repetitions of the AXY8 sequence with the Fourier coefficients $\{f_1 = 0.1, f_2 = f_3 = f_4 = 0\}$ [22]. The ^{13}C layer is modeled by a two-dimensional rectangular grid of 100 ^{13}C nuclear spins, placed at distances of 0.154 nm, corresponding to the next-neighbor distance in the diamond lattice. Our model captures two-spin correlations among the nuclear spins as well as

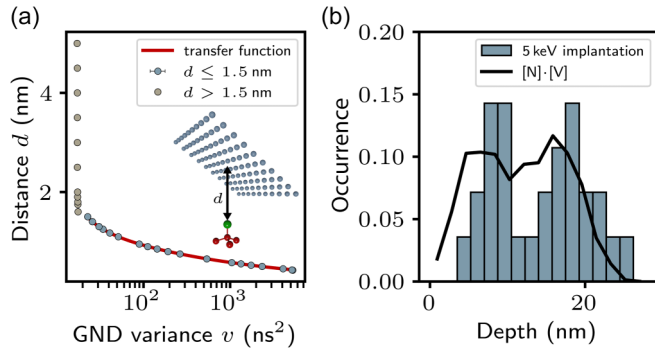


FIG. 5. Distance estimation. (a) Corresponding distances of the extracted variances of the fitted, simulated AXY spectra. The red line (fit) shows the transfer function that is used to estimate the distance of the N-V center to the ^{13}C layer. (b) Depth distribution of a 5 keV implantation. The black solid line shows the product of the simulated vacancy density [V] and nitrogen density [N].

three-spin correlations mediated by the N-V center. A detailed description of the model is found in the Supplemental Material [25].

The simulated spectra are fitted with a generalized normal distribution [27] in the time domain [pulse spacing $\tau = 1/(2\nu)$] and the extracted variances ν for various distances are shown in Fig. 5(a). Beyond 1.5 nm, the signal's amplitude vanishes and the linewidth is solely given by the filter created by the AXY sequence. Therefore, we limit our estimation to distances up to 1.5 nm and phenomenologically fit the obtained variances as a function of distance with the transfer function $d(\nu) = a(\nu - \nu_0)^{-b} + d_0$. We obtain $a = 2.222 \pm 0.017 \text{ nm ns}^{-2b}$, $b = 0.221 \pm 0.005$, $\nu_0 = 14.87 \pm 0.25 \text{ ns}^2$, and $d_0 = 0.0950 \pm 0.0127 \text{ nm}$, with an R^2 of 99.995%.

Applying the same fit routine to the experimental data allows us to then estimate the N-V center- ^{13}C layer distance from the extracted variance via the transfer function. Depend-

ing on the number of observed resonances, the data are fitted with either a single or a double generalized normal distribution, combined with an exponential decay to account for the limited coherence time. Data points that are attributed to sharp nitrogen resonances from imperfect magnetic field alignment are omitted [48].

All extracted variances and estimated distances can be found in the Supplemental Material [25].

APPENDIX C: 5 keV IMPLANTATION COMPARISON

Figure 5(b) shows the depth distribution of a 5 keV $^{15}\text{N}^+$ implantation into a ^{12}C overgrown diamond. The depths are measured for 28 N-V centers via the XY8 sequence [49]. We simulate the implantation with CRYSTAL-TRIM [28] and calculate the product of the vacancy and nitrogen densities as comparison, which is shown by the black solid line in Fig. 5(b). The simulation accurately reflects the profile of the observed distribution but is shifted to smaller depths. This shift can be explained by a slightly different implantation angle, which can alter the channeling effects and thus the final depths. Nevertheless, the simulated product provides a valuable reference for the expected N-V center- ^{13}C layer distance distribution in our samples.

APPENDIX D: ERROR ESTIMATION

The N-V center's normalized fluorescence is obtained by dividing the sum of photon counts during the first $\sim 350 \text{ ns}$ by that of the steady state (recorded for $1.6 \mu\text{s}$) under green ($\lambda = 561 \text{ nm}$) illumination. The measurement error is derived from the corresponding shot noise [50] and propagated using Gaussian error propagation. Unless stated otherwise, all errors reflect the propagated errors of the original fluorescence data. For fit results or quantities derived from fit results, the error is taken as the standard deviation of the fit parameter and propagated accordingly.

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