

Micromotion-based DC sensing using continuously tracked trajectories of dipolar coupled nuclear spins

Ozgur Sahin^{1,*}, Hawraa Al Asadi^{1,*}, Paul M. Schindler^{2,*}, Arjun Pillai¹, Erica Sanchez¹, Matthew Markham³, Mark Elo⁴, Maxwell McAllister¹, Emanuel Druga¹, Christoph Fleckenstein⁵, Marin Bukov^{2,6} and Ashok Ajoy^{1,7}

¹Department of Chemistry, University of California, Berkeley, Berkeley, California 94720, USA

²Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str. 38, 01187 Dresden, Germany

³Element Six Innovation, Fermi Avenue, Harwell Oxford, Didcot, Oxfordshire OX11 0QR, United Kingdom

⁴Tabor Electronics, Inc., Hatasia 9, Nesher 3660301, Israel

⁵Department of Physics, KTH Royal Institute of Technology, SE-106 91 Stockholm, Sweden

⁶Department of Physics, St. Kliment Ohridski University of Sofia, 5 James Bourchier Blvd, 1164 Sofia, Bulgaria

⁷Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA



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We report an experimental approach to excite, stabilize, and continuously track Bloch sphere trajectories of dipolar-coupled nuclear spins in a solid. We demonstrate these capabilities on a model system of hyperpolarized ^{13}C nuclear spins in diamond. We elucidate a method to drive, and preserve, the motion of spins in complex three-dimensional trajectories for over $T_2' > 27$ s even in the presence of interspin coupling. Indeed, without quantum control, interspin interactions lead to rapid spin decay in $T_2^* \approx 1.5$ ms. Furthermore, we show that the motion of the spins can be continuously tracked in three dimensions on the Bloch sphere for over 35 s. During this time the spins complete $>68\,000$ closed precession orbits, exhibiting high stability and robustness against error. Leveraging these long-lived, robust spin trajectories we devise a novel nonequilibrium quantum sensing scheme for DC magnetic fields, based on micromotion dynamics, and without a static counterpart. Sensing here proceeds for the entire T_2' period, orders of magnitude longer than T_2^* , and operates in the dense sensor limit, yielding significant sensitivity improvements. Our results suggest new ways to stabilize and interrogate strongly coupled quantum systems through periodic driving and portend powerful applications of rigid spin orbits in quantum sensing.

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I. INTRODUCTION

Periodically driven quantum systems have attracted enormous interest for their ability to host novel far-from-equilibrium phases of matter [1–5], and for sustaining long-lived, highly stable states wherein absorption of energy can be controllably suppressed [6–8]. Consider, for instance, a network of dipolar-coupled solid-state spins; their interaction drives rapid free induction decay of prepared transverse states in a very short time T_2^* . However, via Floquet prethermalization [8–11], periodically driving the spins can greatly extend the lifetime of these states (from T_2^* to T_2') parametrically with the driving frequency.

However, this approach remains restricted to specific initial states, typically those aligned parallel to the driving field. More generally applicable schemes for stabilization, especially along *arbitrary* axes, remain elusive. In this paper, we propose and demonstrate a strategy to excite and stabilize

closed spin orbits on the Bloch sphere, including those that span large angular excursions away from the driving axis. Our approach exploits novel applications of the eigenstate thermalization hypothesis (ETH) and quantum thermalization [12,13], combined with the engineering of a family of effective Hamiltonians [14] under the simultaneous action of two orthogonal and frequency-separated driving fields. Stable orbits are then excited within the *micromotion* dynamics between these Hamiltonians [15]. In a model system of strongly interacting ^{13}C nuclear spins in diamond with an intrinsic $T_2^* \approx 1.5$ ms [16], we demonstrate the ability to stabilize highly tunable orbits with excursion angles $>15^\circ$ for a lifetime $T_2' > 27$ s. This corresponds to an increase in the spin lifetimes of over 18 000-fold, even in the presence of strong interspin dipolar couplings.

The same method allows us to quasicontinuously track the resulting spin motion in three-dimensions on a Bloch sphere over long periods. Here, we demonstrate the quasicontinuous tracking of spins as they traverse $>68\,000$ cycles of the engineered stabilized orbits. While such state tracking is difficult in many experimental scenarios [17,18], our method makes it easily attainable by: (1) exploiting weak coupling of the nuclear spins to a readout cavity in a manner that imposes no back-action upon them [19]; (2) employing “hyperpolarization,” yielding considerable gains in measured signal to noise [20,21]; and (3) arranging a hierarchical, $>10^5$ -fold

*These authors contributed equally to this work.

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separation of time scales between the rates of cavity-induced spin interrogation (signal sampling) and that of the spin orbiting. Ultimately, this rapid spin tracking unravels the emergence of stable spin orbits, including insights into the underlying dynamical thermalization processes that are key to their rigidity. Leveraging these special features, we demonstrate the ability to excite and control spin trajectories created via dynamically controlled Hamiltonians [22].

Utilizing the stable, long-lived spin trajectories, we introduce a novel nonequilibrium scheme for DC magnetometry with no static counterpart. Our approach exploits the sensitive dependence of trajectory shapes on external fields, effectively translating the DC signal into easier-to-detect AC signals. Exploiting the nonequilibrium (pre)thermal nature of the long-time steady state, sensing can be performed throughout the entire T_2' period, as opposed to conventional approaches which are limited by T_2^* .

Overall, this work expands the toolbox for engineering and stabilizing long-range strongly interacting quantum systems, and via their continuous tracking, heralds diverse applications in quantum sensing [23] and simulation [24]. In particular, we demonstrate that the rigidity and symmetry of motion induced on the spins open new ways to detect weak external fields in the spin environment.

The rest of this paper is organized as follows. Section II presents the basis of our strategy for exciting and stabilizing spin orbits. Section III summarizes our experimental results in producing and controlling long-time rigid polyhedral spin orbits. In Sec. IV, we present how the spin orbits can be used to obtain a novel DC sensor. Technical details underlying these sections are presented in the Appendixes (Appendixes C1–D4).

II. EXCITING AND STABILIZING SPIN TRAJECTORIES

Our experimental system consists of hyperpolarized ^{13}C nuclear spins dipolar-coupled with the interaction Hamiltonian [25], $\mathcal{H}_{\text{dd}} = \sum_{n < m} b_{nm} (3I_{nz}I_{mz} - \mathbf{I}_n \cdot \mathbf{I}_m)$, where $I_{n\alpha}$ refer to spin-1/2 Pauli matrices of nuclear spin n , with $\alpha = \{x, y, z\}$. The coupling strength $b_{nm} = c_{\text{exp}} [3 \cos^2(\beta_{nm}) - 1] / r_{nm}^3$, with the constant $c_{\text{exp}} = \mu_0 \hbar \gamma_n^2 / 4\pi$, and angle $\beta_{nm} = \cos^{-1}(\mathbf{r}_{nm} \cdot \mathbf{B}_0 / r_{nm} B_0)$ of the internuclear vector $\mathbf{r}_{nm}$ in a magnetic field B_0 . At natural abundance ^{13}C concentration, $J = \langle b_{nm} \rangle \approx 0.66 \text{ kHz}$ [11,16], where $\langle \cdot \rangle$ denotes the median over internuclear vector positions. When prepared in a transverse state $\rho_I = I_v = \sum_j I_{j,v}$, where $v = \{x, y\}$, evolution under $\mathcal{H}_{\text{dd}}$ causes the spins to exhibit rapid decay in $T_2^* \approx 1.5 \text{ ms}$ [11]. In addition to $\mathcal{H}_{\text{dd}}$, the spins are also subject to time fluctuating local fields, $\mathcal{H}_z = \sum_j c_j(t) I_{j,z}$, arising primarily from lattice paramagnetic impurities (P1 centers) [26].

One strategy to preserve spin polarization in such transverse states to long times is by Floquet engineering an effective Hamiltonian that conserves the polarization along a $\hat{\mathbf{v}}$ axis to leading order [26]. In Ref. [11], we demonstrated that the resulting transverse lifetimes can exceed $T_2' = 90.9 \text{ s}$ (a $>60\,000$ -fold extension).

This approach, however, only preserves spins along a *single* transverse axis, henceforth labeled $\hat{\mathbf{x}}$ for convenience. Instead, as shown in Fig. 1(c), our goal here is to preserve

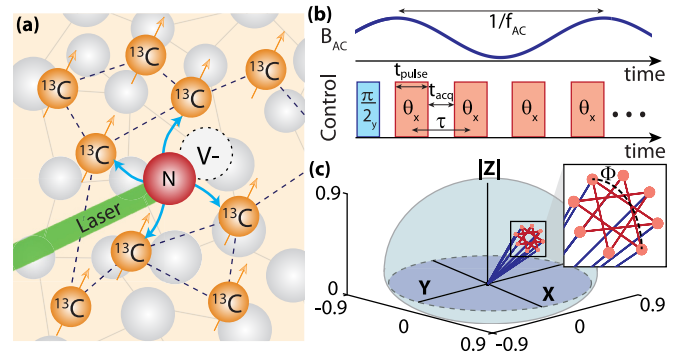


FIG. 1. Concept. (a) *System*: ensemble of dipolar-coupled ^{13}C nuclear spins hyperpolarized by NV centers at 38 mT. (b) *Protocol*: ϑ pulses spin-lock the ^{13}C nuclei at 7 T, which are simultaneously subject to a time-varying field with amplitude B_{AC} and angular frequency $\omega_{\text{AC}} = (2\pi)f_{\text{AC}}$ respectively. Pulse spacing is τ , pulse width is t_{pulse} , and spin precession amplitude and phase are interrogated in t_{acq} windows between pulses. (c) *Experimentally tracked* “eight-point star” spin trajectory on a Bloch sphere during a $\Delta t = 4 \text{ ms}$ window (zoomed in inset). Trajectory remains rigid (stable) for $T_2' \approx 26.4 \text{ s}$, and is continuously tracked for the entire period (see Fig. 12). Extent of the spin excursion on the Bloch sphere is denoted by angle Φ (here $\Phi \approx 11.5^\circ$).

spins in a closed trajectory of arbitrary size, quantified by the excursion angle Φ [shown on Fig. 2(d)] on the Bloch sphere away from $\hat{\mathbf{x}}$. This enables us to design novel sensing protocols, cf. Sec. IV.

A. Floquet protocol

The protocol we employ is described in Fig. 1(b). Spins are subject to a τ -periodic train of spin-locking (SL) ϑ pulses [27,28] (of length t_{pulse} and interpulse spacing τ), while simultaneously being driven by a lower-frequency oscillating (AC) magnetic field along $\hat{\mathbf{z}}$. The Larmor precession of the spins is continuously monitored in windows ($t_{\text{acq}} \sim \tau/2$) between the pulses; the induction signal reports on the precession amplitude and phase in each window. Hyperpolarization, carried out by transferring spin polarization from optically pumped nitrogen vacancy (NV) centers [20,29,30], allows high signal-to-noise readout ($\text{SNR} > 10^3$ per window).

The resulting spin dynamics are most conveniently described by the rotating (dressed) frame Hamiltonian [cf. Appendix D 1],

$$\mathcal{H}_{\text{rot}}(t) = \mathcal{H}_z + \mathcal{H}_{\text{dd}} + \delta I_z + \mathcal{H}_{\text{SL}}(t) + \mathcal{H}_{\text{AC}}(t), \quad (1)$$

where the Hamiltonians corresponding to the orthogonal spin-locking (SL) and the continuous (AC) periodic drives are given by

$$\begin{aligned} \mathcal{H}_{\text{SL}}(t) &= \Omega \Theta(t) I_x, \\ \mathcal{H}_{\text{AC}}(t) &= B_{\text{AC}} \cos(\omega_{\text{AC}} t + \phi_{\text{AC}}) I_z, \end{aligned} \quad (2)$$

where Ω is the Rabi frequency. The time dependence of SL sequence is modeled by a τ -periodic step-function $\Theta(t)$; B_{AC} and ϕ_{AC} refer to the amplitude and phase of the AC field, respectively. The potential term δI_z in Eq. (1) arises from the

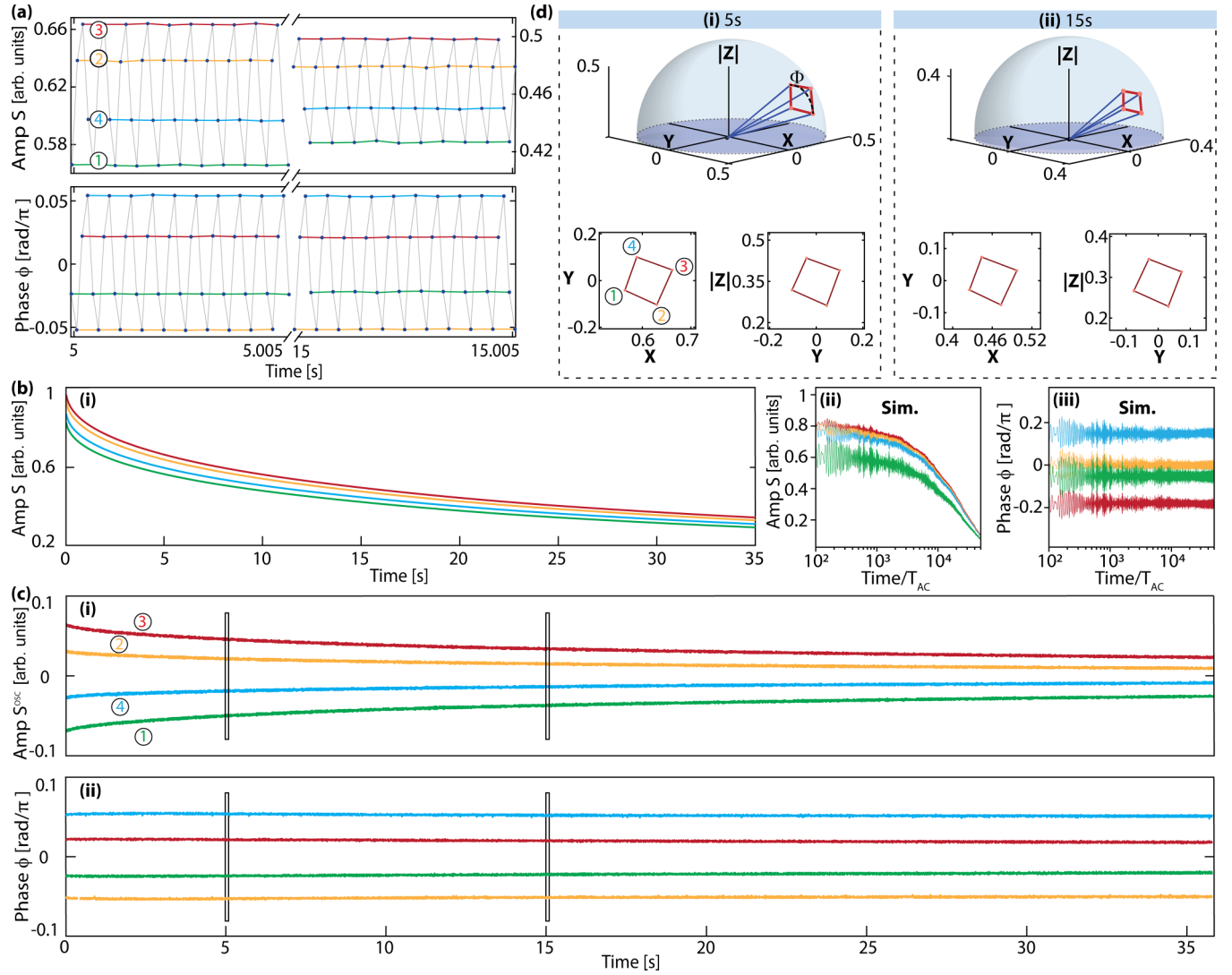


FIG. 2. Long-time continuous tracking of spin evolution. (a) Amplitude S and phase ϕ of spin precession (see Appendix C 1) for two representative $\Delta t = 5\text{ms}$ time windows starting at 5 and 15 s. Here $\vartheta \approx \pi/2$, $\omega_{\text{AC}} = \omega_{\text{res}} = (2\pi)1953.125\text{ Hz}$, matched in period with four pulses [Fig. 1(b)], and shown is an average of 30 experiments. Gray lines connect consecutive oscillating points, and colored lines (manifolds), labeled circled 1–4, connect every fourth point. Individual manifolds display a flat response in each Δt window. (b) (i) Full data for (a) over 35 s, showing signal S with four manifolds separately colored as in panel (a). Decay lifetime is $T_2' \approx 32.5\text{ s}$. (ii), (iii) Numerical simulation of signal S and phase ϕ using a system of $L = 14$ spins (see Appendixes D 1 and E for details of the numerical implementation; simulation parameters are given in Fig. 20). Simulation shows formation of four stable plateaus that ultimately decay to a featureless infinite temperature state (see Appendix D 3). (c) Oscillatory signal (i) amplitude S^{osc} and (ii) phase ϕ extracted from (b) after each of the 273 250 pulses applied for the 35 s period. Slow signal decay is evident, and the four parallel lines in ϕ show that the oscillations persist regularly for many seconds. (d) Bloch sphere representation of spin trajectory in three dimensions (see Appendix C 2), and as 2D projections for two $\Delta t = 5\text{ ms}$ windows starting at (i) $t = 5\text{ s}$ and (ii) 15 s [black narrow rectangles in panel (c)]. Points corresponding to manifolds are marked. Lines join the centroid position of every fourth point in the Δt window. Quadrilateral shaped orbital trajectories are evident; shrinking with time reflects thermalization to an infinite temperature state. Evolution for 35 s in panel (c) corresponds to 68 312 such precessions. We demonstrate the observation of the stable and long-lived spin trajectories resulting from the period matched AC field. Using phase and amplitude data we can reconstruct the spin trajectory on the Bloch sphere.

detuning of the SL pulses from resonance with respect to the Larmor frequency.

On-site random fields $\mathcal{H}_z$ play an insignificant role in dictating the dynamics, and will be ignored for simplicity [31]. Moreover, $\Omega \gg \{B_{\text{AC}}, J\}$ and hence, during the SL pulses, we can also ignore the AC-field-driven evolution and the dipolar dynamics induced by $\mathcal{H}_{\text{dd}}$ to a good approximation (see Appendix D 1). These assumptions allow us to capture the

nonequilibrium dynamics within a simplified Hamiltonian,

$$\mathcal{H}_{\text{rot}}(t) \approx \begin{cases} \delta I_z + \Omega I_x, & t_n \leq t < t_n + t_{\text{pulse}}, \\ \delta I_z + \mathcal{H}_{\text{AC}}(t) + \mathcal{H}_{\text{dd}}, & t_n + t_{\text{pulse}} \leq t < t_{n+1}, \end{cases} \quad (3)$$

where $t_n = n\tau$, $n \in \mathbb{N}$ [see Fig. 1(b)].

In the following, we assume both drives to be commensurate, i.e., such that N -SL pulses are applied in the time the

AC drive completes one period [see Fig. 12(a)] where $N \in \mathbb{N}$. This results in a Hamiltonian $\mathcal{H}_{\text{rot}}(t)$, periodic with frequency ω_{AC} , and allows the use of Floquet theory [32] to analyze the evolution of the system over one AC period $T_{\text{AC}} = 2\pi/\omega_{\text{AC}}$. In particular, we define the unitaries

$$U_{F;k} = \exp(-iT_{\text{AC}}\mathcal{H}_{F;k}), \quad (4)$$

where $\mathcal{H}_{F;k}$ is a discrete family of stroboscopic Floquet Hamiltonians ($k = 1, \dots, N$), obtained by setting the beginning of the stroboscopic frame to be given by the k th SL pulse [2].

While the theoretical analysis is made complex due to the simultaneous application of continuous and kicked drives, it is analytically feasible when ϑ is close to $2\pi/N$, i.e., $\vartheta = 2\pi/N + \delta\vartheta$ with $\delta\vartheta \ll 1$. We employ the Floquet-Magnus expansion [33] in the SL toggling frame over one AC drive period, to expand $\mathcal{H}_{F;k}$ in the inverse frequency ω_{AC}^{-1} . In the high-frequency regime $\omega_{\text{AC}} \gg \{J, \Omega, B_{\text{AC}}, \delta\}$, the dynamics are governed by the time-independent effective Hamiltonian (see Appendix D 2)

$$\begin{aligned} \frac{t_{\text{acq}}}{\tau} \tilde{\mathcal{H}}_{F;k}^{(0)} &= \tilde{\mathbf{w}}_k(\alpha, \phi_{\text{AC}}) \cdot \mathbf{I} \\ &+ (1 - 3\sin^2\alpha) \sum_{n < m} b_{nm} \left(\frac{3}{2} \tilde{\mathcal{H}}_{\text{ff}} - \mathbf{I}_n \cdot \mathbf{I}_m \right), \end{aligned} \quad (5)$$

with the flip-flop term $\tilde{\mathcal{H}}_{\text{ff}} = I_{jz}I_{kz} + I_{jy}I_{ky}$, and $\alpha(\vartheta) = \arctan(\delta/\Omega)$ (here the tildes denote operators transformed under the change of basis mapping $(\cos(\alpha)I_x + \sin(\alpha)I_z) \rightarrow I_x$, see Appendix D 2). An effective static field arises,

$$\begin{aligned} \tilde{\mathbf{w}}_k(\alpha, \phi_{\text{AC}}) &= (\delta \sin \alpha + \delta\vartheta/\tau) \hat{\mathbf{x}} + B_{\text{AC}} \cos \alpha \sum_{n=1}^N (-\sin n\vartheta \hat{\mathbf{y}} \\ &+ \cos n\vartheta \hat{\mathbf{z}}) f_{n+k}(\phi_{\text{AC}}), \end{aligned} \quad (6)$$

which changes direction conditioned on k , since $f_{n+k}(\phi_{\text{AC}}) = \int_{t_n+t_{\text{pulse}}}^{t_{n+1}} \sin(\frac{2\pi t}{T_{\text{AC}}} + \phi_{\text{AC}}) \frac{dt}{t_{\text{acq}}}$; $\tilde{\mathbf{w}}_k$ elucidates the *micromotion* of the spins as they are successively kicked by the SL pulses. Importantly, the interaction term in Eq. (5) remains independent of the stroboscopic frame index k .

B. Magnetized prethermal plateau without a quasiconservation law

Equation (5) is a key result of this work. Within the duration of the parametrically long-lived prethermal plateau, the spins thermalize under the nonintegrable Hamiltonian $\tilde{\mathcal{H}}_{F;k}^{(0)}$. Hence, we anticipate the formation of a prethermal state effectively captured by density matrix $\tilde{\rho}_F \propto \exp(-\beta \tilde{\mathcal{H}}_{F;k}^{(0)})$ with the inverse temperature β set by the energy density of the initial state, in a similar manner to ETH [12,13]. In conventional SL, the leading-order Floquet Hamiltonian commutes with the polarization $\mathcal{I}_x$ in the SL direction, and this gives rise to a long-lived prethermally magnetized state. By contrast, in our system this quasiconservation law is broken by the extra commensurate kicks. Naïvely, one would thus expect a thermal state at infinite temperature, without any leftover polarization. However, it turns out that by selecting a detuning $\delta \neq 0$ (or $\delta\vartheta \neq 0$), the initial state $\tilde{\rho}_I \propto \mathcal{I}_x$ features a small but nonvanishing energy density, $\text{Tr}(\tilde{\rho}_I \tilde{\mathcal{H}}_{F;k}^{(0)}) \propto (\delta + \delta\vartheta/\tau)$, which

leads to a stable long-lived prethermal plateau with a finite temperature $\beta > 0$. Hence, for $\beta J \ll 1$, we have $\tilde{\rho}_F \propto \tilde{\mathcal{H}}_{F;k}^{(0)}$. We emphasize that the physics of this state differs substantially compared to the spin-locked prethermal state, where $\tilde{\rho}_I \propto \exp(-\mu_x \mathcal{I}_x)$ for some chemical potential μ_x .

From Eq. (5), the prethermal state carries both single-body (polarization) terms and “dipolar order” [34–37], where our realization of the latter stands in contrast to previous methods using the Jeener-Broekaert sequence [35,38,39]. Let us denote by $\mathcal{I}_v$ the polarization components in the rotating frame. Then, in the prethermal plateau, the expectation value of the polarization $\mathbf{M} = (\mathcal{I}_x, \mathcal{I}_y, \mathcal{I}_z)$ is given by $\mathbf{M} = \text{Tr}[\mathbf{I} \tilde{\rho}_F] \propto \tilde{\mathbf{w}}_k$. It points in different directions for every k value within the drive period T_{AC} . Since the micromotion dynamics are continuous and cyclic with the drive period, the polarization vector will follow periodic excursions on the Bloch sphere. The extent of these excursions can be quantified by the angle Φ [see Fig. 2(d)]. Importantly, since the Floquet Hamiltonians $\tilde{\mathcal{H}}_{F;k}^{(0)}$ are related to one another by a static change of basis, and the ϑ pulses just serve to cause micromotion between them, there is *no* transient in the dynamics of the thermalized spins between successive pulses.

As a consequence, when observed at times $t_n + t_{\text{pulse}}$, the polarization features N metastable plateaus ($N = 4$ in Fig. 2). From the perspective of Floquet theory, the observed plateaus can be interpreted as a single plateau, periodically displaced in time by the micromotion dynamics. The lifetime of any of the plateaus is therefore determined by the intrinsic T'_2 lifetime set by SL prethermalization; in our long-range three-dimensional (3D) system, the plateau lifetimes depend parametrically as $T'_2 \sim (J\tau)^{-2}$ [14]. This forms the basis for exciting and preserving large closed orbits on the Bloch sphere in our experiment. To the best of our knowledge, this is the first time that the observation of micromotion, combined with quantum thermalization, has been proposed as a stabilization mechanism for spin orbits in the absence of a quasiconserved quantity.

III. CONTINUOUSLY TRACKED PRETHERMAL SPIN TRAJECTORIES

A. Methodology

An important practical achievement of this paper is the ability to continuously *track* the rotating-frame spin trajectories in three dimensions on the Bloch sphere.

Typically, spin trajectory tracking is challenging because it requires: (1) the ability to interrogate the spins without perturbing them and (2) that state interrogation can occur simultaneously with the control field $\mathcal{H}_{\text{AC}}$. To make such tracking viable, we employ a series of special features in our system: (i) weak spin-cavity coupling that permits non-destructive measurements via RF induction, (ii) a hierarchical $> 10^5$ -fold timescale separation between the rate at which the induction signal is sampled and cyclic orbital motion (see Table I), and (iii) hyperpolarization, which yields high single-shot readout SNR.

We employ quadrature information from each acquisition window following a SL pulse to extract transverse spin projections in the rotating (dressed) frame, $\mathcal{I}_x(t_j) = S(t_j) \cos \varphi(t_j)$, and $\mathcal{I}_y(t_j) = S(t_j) \sin \varphi(t_j)$, where S and φ denote the ampli-

TABLE I. Timescale separation between parameters of spin evolution and cavity interrogation (see Fig. 8). Typical values employed in experiments are listed in third column, while the last column shows them normalized with respect to B_{AC} . There is a $\sim 10^5$ -fold separation between ω and ω_L^s that allows quasicontinuous sampling.

Parameter	Particular	Value	Norm. Value
$(T_2')^{-1}$	Prethermal decay rate	~ 10 mHz	10^{-2}
$\gamma_{B_{AC}}$	Strength of AC field	0.01–1 kHz	1
$\omega/(2\pi)$	Dressed frame res. freq.	0.1–20 kHz	10–2000
$\omega^s/(2\pi)$	Trajectory sampling rate	1 kHz–1 MHz	10^2 – 10^5
$\omega_L/(2\pi)$	Cavity resonance frequency	75 MHz	7.5×10^6
$\omega_L^s/(2\pi)$	Sampling of cavity evolution	1 GHz	10^8

tude and phase in this frame, and variable t_j captures quasicontinuous sampling at rate $\omega^s = (2\pi)\tau^{-1}$. In practice, obtaining $\varphi(t)$ requires removing the trivial phase accrued due to Larmor precession during each SL pulse, equivalent to a transformation from laboratory frame to rotating frame (see Fig. 9).

Since the spins undergo very little decay during successive cavity interrogation windows, $(\omega^s T_2')^{-1} \ll 1$, it is possible to estimate the magnitude of the $I_z(t_j)$ component through a unitarity constraint, $|I_z(t_j)| = [\mathcal{N}^2(t_j) - I_x^2(t_j) - I_y^2(t_j)]^{1/2}$; here $\mathcal{N}(t)$ refers to the instantaneous norm of the polarization vector. This allows us to track and plot the total polarization $M(t) = \{I_x(t), I_y(t), I_z(t)\}$ (sampled at ω^s) on a Bloch hemisphere for long times, yielding a quasi real-time 3D map of their orbital trajectories [Fig. 1(c)]. The methodology we employ is elucidated in detail in Appendix C 1.

B. Experimental observation

In Fig. 2 we demonstrate the ability to excite, stabilize, and track spin trajectories on the Bloch sphere for long periods (here 35 s). Figure 2(a) shows the tracked amplitude S and phase φ employing the protocol in Fig. 1(b) with $\vartheta = \pi/2$ and $B_{AC} = 185 \mu\text{T}$, shown for two $\Delta t = 5$ ms windows starting at $t = 5$ s and $t = 15$ s, respectively. Evidently, the amplitude decay during the ~ 10 s intervening period is relatively small [cf. left and right y axes in Fig. 2(a)]. More striking, perhaps, is the regularity of the four-point periodic oscillations (gray lines), reflecting the micromotion dynamics. To reveal this more clearly and highlight its stability, in Fig. 2(a) we join every fourth point by lines of different colors and label these manifolds as ①–④.

The full signal S up to $t = 35$ s is shown in Fig. 2(b(i)). We use the same color convention as in Fig. 2(a), but ignore the gray line joining successive points. Spin prethermalization causes each signal manifold to decay with a stretched exponential profile $\exp[-(t/T_2')^{1/2}]$ with $T_2' \approx 33$ s, cf. Figure 2(b(i)) (see also Ref. [40]). Figures 2(b(ii)) and 2(b(iii)) shows full numerical simulations of S and φ following Eq. (3), employing a system of $L = 14$ spins and averaging over multiple disorder manifestations. It is possible to discern four stable plateaus (colored) that undergo slow prethermal decays to infinite temperature. The small size of the simulations results in larger fluctuations compared to the experimental data in Fig. 2(b(i)).

To highlight the micromotion dynamics, it is helpful to separate the effect of the background decay. Indeed, the

prethermal decay profile, S^{dec} , can be extracted by smoothing S with a moving average filter. Then the micromotion is captured by the oscillatory part $S^{\text{osc}}(t) = S(t) - S^{\text{dec}}(t)$, and is shown in Fig. 2(c(i)) for $t = 35$ s. Corresponding phase $\varphi(t)$ signal is shown in Fig. 2(c(ii)). Data here comprises 273 250 spin-lock pulses and the completion of 68 312 four-point periodic orbits. High stability of the micromotion is reflected in the observation that the four amplitude and phase manifolds remain separate and almost parallel over the entire 35 s period. We note that the stability persists in spite of ever present RF inhomogeneity in the SL pulses (see Appendixes C 5 a and C 5 b for a full discussion of the origin of this enhanced robustness). While finite computer memory limitations impose restrictions to the total observation period in current experiments, from the data in Fig. 2(c), we estimate that the stable dynamics should persist up to $t > 200$ s.

The high stability makes our spin tracking method (Appendix C 2) viable and carries a low degree of error. This is reflected in Fig. 2(d) where we reconstruct the full 3D dynamics for two $\Delta t = 5$ ms windows starting at (i) $t = 5$ s and (ii) $t = 15$ s [narrow rectangular windows in Fig. 2(c)] on a half Bloch sphere. Spins trace a four-point orbit on the Bloch sphere that resemble quadrilaterals; we will refer to the trajectories as being “squarelike” for convenience. Comparison of the two panels in Fig. 2(d) reflects the apparent shrinking of the orbits due to prethermal decay during the intervening period between these Δt windows. Lower panels in Fig. 2(d) show corresponding 2D projections in the $\hat{y}$ - $\hat{z}$ and $\hat{x}$ - $\hat{y}$ planes. Plotted are ≈ 10 points per manifold [labeled as in Fig. 2(c)] that correspond to the extracted spin positions in the $\Delta t = 5$ ms window; due to high SNR and stability, the points here are essentially overlapping. Red solid lines join the points in both the 2D and 3D representations. The excursion angle in Fig. 2(d(i)) is $\Phi = 15.89^\circ$, and we ascribe the overall tilt of the trajectory away from $\hat{x}$ to detuning of the SL pulses. This is also supported by the fact that the $\hat{x}$ - $\hat{z}$ projections trace arcs (see Appendix C 4 and Fig. 13).

Panels in Fig. 2(d), corresponding to small $\Delta t = 5$ ms windows, only represent a tiny sliver ($\sim 10^{-4}$ times) of the full dynamics tracked in Fig. 2(c). Indeed, it is possible to plot the resulting trajectory for the entire 35 s period, but the motion is most conveniently captured by focusing on a single projection. Figure 12(c) shows the results of such a visualization, where we cast the $\hat{y}$ - $\hat{z}$ projection on a horizontal plane and time on the vertical axis.

C. Probing the properties of the stabilized shapes

To probe the relative stability of the trajectories with the excursion angle, we excite squarelike trajectories (cf. Fig. 3) of varying sizes by changing the AC amplitude $|B_{AC}|$. Figure 3(a) shows two representative tracked trajectories with $B_{AC} = 42 \mu\text{T}$ and $210 \mu\text{T}$, respectively. Figure 3(b) shows the $\hat{y}$ - $\hat{z}$ projection averaged over a $\Delta t = 15$ ms window centered at $t = 3$ s for a range of B_{AC} values [see colorbar in Fig. 3(e)]. The upper limit of B_{AC} values employed here is due to heating in the AC field coil. In Fig. 3(c), we extract the corresponding (average) radius ℓ of the “square” trajectories (blue points) and corresponding excursion angles

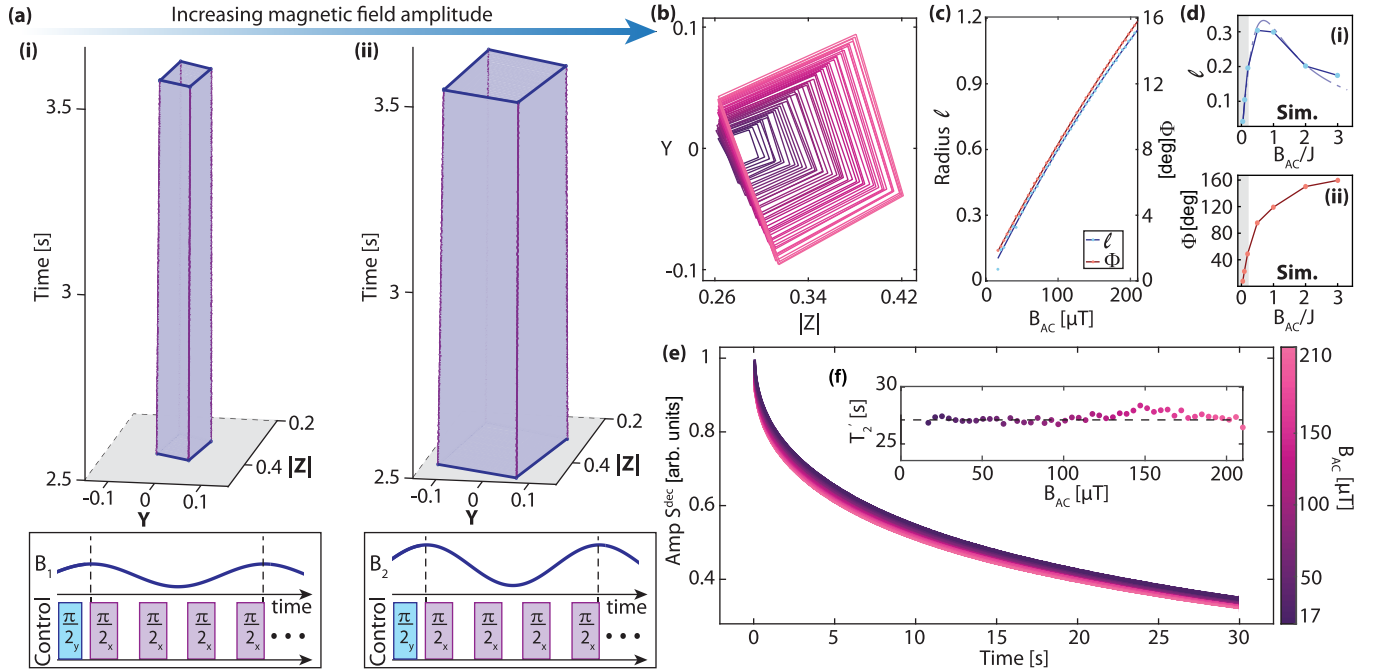


FIG. 3. Stabilized spin trajectories at high amplitudes. (a) *Tracked spin trajectory* for a squarelike trajectory (see Fig. 12) shown for a $\Delta t = 1$ s section under two representative fields: (i) $B_1 \approx 42 \mu\text{T}$ and (ii) $B_2 \approx 210 \mu\text{T}$ (lower panels show schematic control sequences). Excursion angles are $\Phi \approx 4.05^\circ$ and $\Phi \approx 15.89^\circ$ respectively. (b) *Top-view* (in $\hat{y}$ - $\hat{z}$ plane) of the spin trajectories with varying B_{AC} [see colorbar in panel (e)]. Curves here are an average over 50 slices. (c) Corresponding increase in excursion angle Φ (red points) and radius ℓ (blue points) with B_{AC} . Solid lines are spline fit guides to the eye. (d) Numerically simulated scaling of (i) radius ℓ and (ii) excursion angle Φ as a function of B_{AC}/J (see Fig. 21 for full data). In the former, the dashed line follows exact analytical predictions (see Appendix D 3). The shaded regions here correspond to the experimentally relevant regime of B_{AC} values in panel (c). (e) *Spin trajectory decay profiles* S^{dec} under different amplitudes B_{AC} (see colorbar). Shown are smoothed raw amplitude S curves (see Appendix B for discussion). (f) T_2' lifetimes extracted from stretched exponential fits to profiles in panel (e). Data reveals corresponding lifetimes $T_2' \approx 27.0 \pm 0.2$ s (dashed line), approximately independent of B_{AC} . In agreement with theory predictions, we are able to engineer and stabilize spin trajectories with relatively large deviations from the $\hat{x}$ axis (see Appendix B).

Φ (red points). Ultimately, at $B_{AC} = 210 \mu\text{T}$ we estimate an excursion angle of $\Phi = 15.89^\circ$.

We can derive approximate analytical expressions that quantify the dependence of the radius ℓ and the excursion angle Φ on the system parameters. As already mentioned, the measured prethermal polygon shapes can be characterized by the radius ℓ of the micromotion circle on which the edges of the shapes lie, and the excursion angle Φ away from the spin-lock axis. A more detailed analysis (see Appendix D 3) shows that

$$\ell \propto \frac{\varepsilon_k B_{AC}}{h_{dd}^2 + |\mathbf{w}_k|^2}, \quad (7)$$

where $\varepsilon_k = \text{Tr}\{\tilde{\rho}_0 \tilde{\mathcal{H}}_{F,k}^{(0)}\}/L$ is the energy density in the prethermal plateau; the dipole-dipole interaction strength is given by $h_{dd}^2 = 2(3 \sin^2 \alpha - 1)^2 \sum_{n < m} b_{nm}^2 / L \propto J^2$, and $\mathbf{w}_k$ is the vector traced by the micromotion defined in Eq. (6). Decomposing $\mathbf{w}_k = \mathbf{w}_{k,\parallel} + \mathbf{w}_{k,\perp}$ in components parallel and perpendicular to the polygonal base of the shape, we define the excursion angle implicitly via the relation

$$\tan\left(\frac{\Phi}{2}\right) = \frac{\ell}{|\mathbf{w}_{k,\perp}|} \propto B_{AC}. \quad (8)$$

Both quantities ℓ and Φ are thus controlled by B_{AC} , and can be used to sense small changes in the presence of

additional external AC and DC fields. In Fig. 3(d)(i) and 3(d)(ii) show that our analytical expressions yield excellent qualitative agreement with numerical simulations, respectively (see also Appendix E). It is important to note that the shapes constitute a prethermal phenomenon and, as such, are expected to decay at sufficiently long times. Figure 3(e) shows the corresponding S^{dec} decay profiles obtained by smoothing raw amplitude S curves (see colorbar for B_{AC} values). Overlapping curves in Fig. 3(e) indicate that the respective decay rates are approximately independent of the AC field amplitude B_{AC} . This is clearer in Fig. 3(f), where we extract the corresponding stretched exponential decay constants T_2' . The data reveals $T_2' \approx 27.0 \pm 0.2$ s (horizontal dashed line) is independent of B_{AC} to a good approximation. These conclusions are supported by our numerical simulation in Fig. 21 where we extract T_2' as a function of B_{AC}/J for an $L = 14$ system. These results demonstrate the ability to stabilize, via inter-spin interactions, large spin trajectories on the Bloch sphere while also continuously tracking the resulting motion—a key achievement of this paper.

D. Robustness and control of spin trajectories

Let us emphasize that our procedure is not limited to engineering squarelike trajectories but can be used to engineer

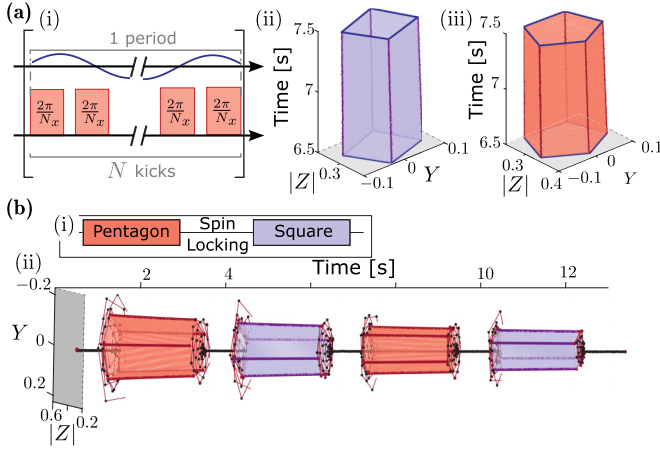


FIG. 4. Control and stability of spin trajectories. (a) (i) *Schematic*: Pulse sequence to create a N -polygon experimental observation of square (ii) and pentagon shapes (iii). (b) (i) *Schematic*: creation and stabilization of square and pentagon shaped trajectories. (ii) *Multiple-polyhedron spin trajectory* showing four periods of 2 s long square and pentagon trajectories (separated by $\tau_* = 1$ s). See Appendix C 5 c (Fig. 16) for comparison of decay profile with a single square trajectory over the entire period. The spin trajectories can be dynamically controlled within the prethermal plateau with little to no loss in signal and lifetime.

also other trajectories. For concreteness, to obtain a n -polygon the AC-period must be commensurate k kicks, with each kick having a kick angle close to $2\pi/n$. For example, using $n = 4$ or $n = 5$ we can obtain squares or pentagons see Fig. 4(a), respectively. In Appendix C 4, we detail the engineering of more complex shapes. Engineering the properties of the robust spin trajectory is a fundamental step; yet, this is still insufficient for most practical applications without having the ability to dynamically control and modify these properties within the prethermal lifetime. In general, control of nonequilibrium systems in the prethermal plateau is a delicate task; arbitrary control will interfere with the periodicity of the drive, potentially diminishing the lifetime or even compromising the stability of the plateau altogether. In Fig. 4, we experimentally demonstrate the ability to robustly control the properties of the system during the prethermal plateau. In particular, we consider the sequence in Fig. 4(a) of opening and closing the trajectories. Starting from standard spin-locking with a single prethermal plateau we can open and close spin trajectories with different numbers of edges; see Fig. 4(b). While opening and closing the shapes results in short-lived transients, the entire control happens with little to no losses in amplitude; see Fig. 4(b) (see also Fig. 16). In the following, we will build on the properties of these robust nonequilibrium trajectories to construct protocols for improved quantum sensing.

IV. APPLICATIONS: NONEQUILIBRIUM QUANTUM SENSING OF DC FIELDS

We now describe applications of the driven spin orbits toward the quantum sensing of DC and low-frequency magnetic fields, important in several contexts [41,42]. Traditional DC magnetometry methods are significantly less sensitive than

AC magnetometry because echo-based techniques in the latter allows the ability to suppress noise on the sensor. Indeed, discrepancy between both cases is particularly large when (1) the ratio T_2/T_2^* is large, as is the case in most experimental situations [43], (2) for sensing in the high-density limit, wherein T_2^* is dominated by intersensor interaction [44], and (3) when the initialization t_p (or readout) dead times, are significantly larger than T_2^* . This is a common situation when considering dense sensor ensembles [45]. For instance, for diamond crystals with 10ppm NV centers and isotopically enriched in ^{12}C , the NV center coherences follow $T_2^* \sim 800$ ns, $T_2 \sim 10$ μs , and $t_p \sim 1$ μs [45]. Increasing sensor density normally deleteriously affects T_2 ; as a result most quantum sensing is traditionally performed in limit of (dilute) noninteracting spins, and increasing sensor constitutes an important frontier for sensitivity improvement [46].

Engineered spin orbits, and exploiting sensor prethermalization, provide a conceptually novel means to overcome these challenges. To summarize, we propose here to sense a DC magnetic field, B_{DC} , indirectly through the response to an AC magnetic field. Figure 5(a) illustrates the concept: the spins are driven into an orbit via an AC field; however, the orbit size is strongly influenced by the presence of B_{DC} . Importantly, the sensing can work efficiently here because the interrogation of the DC field can be sustained till T_2' , significantly larger than T_2^* , and in the dense sensor limit. Indeed, T_2' is generally even larger than the echo-lifetime T_2 , which instead is not immune to interspin interaction [47]. In the limit of rapid pulsing, the spin-locking lifetime $T_2' = T_{1\rho}$ can potentially be a significant proportion of T_1 [48,49]. The hyperpolarized ^{13}C nuclei here are particularly well-suited to illustrate the advantages possible through this approach, since $T_2'/T_2^* > 10^4$, and hyperpolarization time $t_p/T_2' \sim 1$. However, one can argue that this methodology is quite generally applicable in dense sensor ensembles.

To now detail specifically the operation of the sensor, consider that for an $\hat{x}$ -polarized initial state, $\tilde{\rho}_0 \propto (1 + \mu I_x)$, Eq. (7) postulates that the radius ℓ of the shape depends strongly on the detuning $\delta = \omega_{\text{SL}} - \omega_L$ between the spin-locking and Larmor frequencies, via the energy density:

$$\varepsilon_k = \mu \left| \frac{\delta^2}{\sqrt{\delta^2 + \Omega^2}} + \frac{\delta \vartheta}{\tau} \right|. \quad (9)$$

This can be leveraged to construct a nonequilibrium protocol for DC sensing exploiting key ingredients: (i) prethermalization to engineered effective Hamiltonian $\rho \sim \tilde{H}_{F,k}^{(0)}$, which requires an ergodic system and a nonequilibrium Floquet drive; (ii) observable micromotion dynamics on a controllably long prethermal timescale, to induce stable long-lived oscillations within the trajectory; (iii) high sensitivity of the trajectory axis to small changes in the detuning parameter δ .

The relevance of the individual steps is as follows. Due to (i) each of the many spin ensembles ρ_k is in a steady state and therefore fully robust against intrinsic disorder. The key property of micromotion, step (ii), is that it translates between different prethermal plateaus, hence allowing to transform the DC signal into an AC response. We stress that an analogous scheme cannot be realized in an equilibrium setting, since any AC response is directly associated with a nonequilibrium

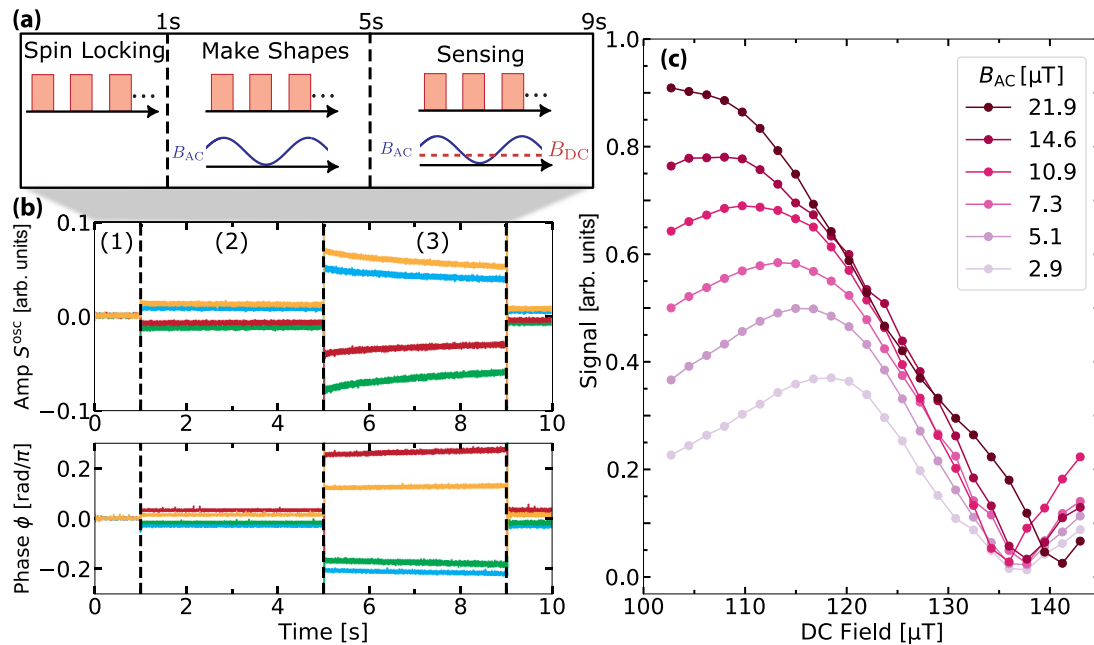


FIG. 5. DC Sensor. (a) Sketch of the sensing protocol: The sensor is initialized in block (1) and (2) which spin lock the spins along the $\hat{x}$ axis and create the shape in the linear regime of the sensor, respectively; sensing is performed in block (3). (b) Observed oscillating amplitude S^{osc} (top) and phase ϕ (bottom) for protocol in A. (c) Dependence of the signal on the applied DC Voltage for different AC field strengths $B_{AC} = 40\text{--}300\text{ mV}_{pp}$ (light to dark). Signal is extracted as the peak amplitude in the power spectrum $P(f)$ around $f = f_{AC}$. The high sensitivity of the spin trajectories to the applied DC Voltage makes them particularly suitable for application as a DC magnetometer; the dynamical range of the sensor can be tuned within a factor of ~ 2 by varying the applied AC field.

setup. We also emphasize that, via points (i) and (ii), our protocol differs significantly from previous state-of-the-art sensing protocols which also rely on unitary evolution.

Figure 5(a) walks through the experimental scheme in a step-wise manner. Initially, we spin-lock the spins along the $\hat{x}$ direction until $t = 1\text{ s}$ (Block 1), resulting in prethermalization along the $\hat{x}$ axis. Subsequently, the resonant AC drive is turned on which prethermalizes the system to $\rho \sim \mathcal{H}_{F,k}^{(0)}$ until $t = 5\text{ s}$ is reached (Block 2). Blocks 1 and 2 serve as the initialization phase for the sensor. At $t = 5\text{ s}$, we turn on a small ($\sim 120\text{ }\mu T$), static magnetic field B_{DC} along the $\hat{z}$ axis until $t = 9\text{ s}$ (Block 3). Figure 5(b) shows the amplitude and phase data taken under this scheme for 10 s. In Block 1, both the amplitude and phase data show a single line, indicating that the system is indeed spin-locked along the $\hat{x}$ axis. In Block 2, the trend on the amplitude and phase data follow Figs. 2(b) and 2(c) closely, displaying four parallel manifolds. The data in Block 3 qualitatively resembles that in Block 2, featuring four distinct plateaus. However, there is a notable quantitative change in the amplitude and phase data in Block 3 compared to Block 2. Importantly, this shows that a state which has prethermalized into the $\rho \sim \mathcal{H}_{F,k}^{(0)}$ manifolds, reacts to DC fields mainly by changing the intensity of the AC field oscillations. This property makes it suitable for application as a DC magnetometer.

Figure 5(c) shows the oscillation magnitude for the amplitude data in Block 3 as a function of the DC field at fixed detuning δ . The data reveals that the oscillation amplitude is strongly dependent on the magnitude of the DC field, especially in a dynamic range of $\sim 15\text{ }\mu T$ near the minimum of the signal. This susceptibility to external DC

fields, enables us to exploit the spin trajectories as DC sensor; from Fig. 5(c) we can extract a sensitivity of $\sim 26 \pm 3.6\text{ nT}/\sqrt{\text{Hz}}$; see Appendix A. In particular, this outperforms the Ramsey sensitivity for the same setup ($\sim 480\text{ nT}/\sqrt{\text{Hz}}$, see Appendix A) by more than an order of magnitude—a reflection of the gains possible in the high-sensor density, high overhead-time regime as operational here. The DC sensing here is also in a high-bias field environment (7 T); the sensing here compares very favorably to most DC magnetometer technologies at these high-field settings [50].

With respect to absolute (bias-field-agnostic) sensitivity, we note that NV electron-based DC magnetometers perform better, with sub-picotesla operation possible [51], primarily because of the lower gyromagnetic ratio for the ^{13}C nuclei. However, our goal was to demonstrate a new concept of T_2' -limited DC sensing in dense ensembles, and while our experiments are on ^{13}C nuclear spins as a proof-of-concept, a similar protocol is directly extensible to NV center ensembles.

We can tune the linear range of the sensor by controlling the detuning frequency δ as a knob for attenuating or increasing the DC signal by a controllable amount; hereby, the main limiting factor in precision is the frequency fidelity of the RF pulse generator. Similarly, it is possible to control the dynamic range to an extent by changing the AC magnetic field amplitude B_{AC} ; see Fig. 5(c).

V. CONCLUSIONS AND OUTLOOK

Our work has unexplored potential to be extended in diverse directions, both fundamental and applied. Our novel nonequilibrium DC sensing scheme demonstrates that

micromotion can be turned into a powerful tool in quantum sensing, rather than being a mere artifact. The intricate properties of the spin orbits imprinted by micromotion, offer exciting new avenues for quantum sensing. In particular, we expect that correlations between different plateaus may be exploited for correlated sensing. Moreover, the symmetry imposed on the spin orbits (Fig. 12) may allow higher selectivity for specific $\mathcal{H}_{AC}$ frequencies of interest over noise and other frequencies, which do not produce any symmetry of the spin motion.

Furthermore, our method may make *dense* spin ensembles practical for these use cases by exploiting thermalization-controlled micromotion dynamics. This would substantially expand the scope of quantum sensing beyond the conventional dilute (single-spin) limit [46,52]. Beyond high-field magnetometry with ^{13}C nuclear spins [50], we anticipate that this approach can be extended to NV center ensembles [53] and polar molecules [54] where analogous driving schemes may be implemented. Finally, spin orbits under a gradient may serve as an encoding strategy for hyperpolarized MR imaging [55].

While our experiments have focused on hyperpolarized ^{13}C nuclear spins in diamond, organic molecules which host triplet photoexcitable electrons [56–58] might prove even more compelling. For instance, Ref. [59] measured the T_1 lifetimes of optically hyperpolarized ^1H nuclear spins in pentacene to be >100 hr at 77 K.

On the fundamental side, the experiments here provide a new way to interrogate thermalization dynamics in nonequilibrium interacting quantum systems [60]. Moreover, the use of Floquet prethermal plateaus has remained largely unexplored in the context of state manipulation thus far. The robustness of the spin orbits to errors (Appendixes C 5 a and C 5 b) suggests practical applications for quantum gates without the need to decouple interactions between the spins.

Finally, the principle for long-time quasicontinuous spin tracking developed here is generally applicable to wide classes of control fields $\mathcal{H}_c(t)$. As such, this portends impactful applications in quantum feedback [61,62], Hamiltonian estimation [63], and the design of optimal control protocols [64–67].

ACKNOWLEDGMENTS

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APPENDIX A: METHOD FOR CALCULATING DC FIELD SENSITIVITY

In this Appendix, we describe in detail how we compute the dc sensitivity reported on in the main text; see Sec. IV.

First, we need to estimate a conversion factor between voltage and magnetic field as the DC and AC fields are generated by a coil driven by two signal generators (one for AC and the other for DC) at constant voltage mode. To this end, we exploit the fact that the oscillation intensities are minimized when the pulse frequency is at resonance with the Larmor frequency of the ^{13}C nuclei. We sweep the frequency of the NMR pulses, once with the coil not being driven and then with it being driven by a voltage of 800 mV. We pick the two frequencies where the oscillation intensities are at a minimum and then estimate the magnetic field as

$$B = \frac{\Delta f}{\gamma}, \quad (\text{A1})$$

where Δf is the difference in frequencies, and γ is the gyromagnetic ratio of the ^{13}C nuclei. From here, we estimate the conversion factor to be ~ 146 nT/mV.

Following Ref. [50], we can estimate the sensitivity as the minimum change in the external field that creates a response in the signal on the order of the variance of the noise level in the frequency domain. For concreteness we compute the sensitivity for the dataset with the greatest dynamic range (i.e., $B_{AC} \approx 20$ μT); see Fig. 5(c). The uncertainty of the signal along the y axis is extracted from the noise level in the power spectrum of each data point in Fig. 5(c). We can then estimate the uncertainty in the sensed magnetic field by dividing the uncertainty in the signal by the linear slope of the data in the linear regime. Finally, we normalize our data (4 s acquisition plus 40 s dead time for hyperpolarization) to 1 s, to find a sensitivity for our nonequilibrium sensing scheme of $\sim 26 \pm 3.6$ nT/ $\sqrt{\text{Hz}}$ as reported in the main text. We refer the reader to Ref. [50] for a more detailed explanation of this method.

For Ramsey sensitivity estimation, we follow a slightly modified protocol. We take one readout shot of the raw data (see Appendix B 3) that is ~ 33 μs long and Fourier transform it to find the behavior around 20 MHz in the frequency domain. The resulting plot looks like a sinc function centered around 20 MHz. We then fit a line through the linear portion of the central peak of the sinc function and the slope for that fit acts as the conversion factor from the signal to frequency in units of MHz which is later converted to magnetic field by dividing by the gyromagnetic ratio γ of the ^{13}C nuclei. The noise floor is estimated by the standard deviation in the signal at 95% CI level between 450 and 460 MHz where no peaks are present. The minimum detectable magnetic field for this single shot is then estimated to be this noise level converted into magnetic field by the aforementioned conversion factor.

In a sensing scheme that has an exponential decay profile and a small dead time compared to the sensing time, the maximum sensitivity is achieved when the sensor is operated for a time $T_2^*/2$ [23]. We additionally assume that the minimum detectable field is inversely proportional to $\sqrt{N}$ where N is the number of shots in a given experiment. Using these two assumptions, we can compute the sensitivity for the Ramsey scheme of as follows: N can be estimated simply by $T_{op}/t_{shot} \sim 7$ where $t_{shot} = 108$ μs (63 μs pulse time, 33 μs readout time, and 11 μs dead time) is the time for a single shot, and T_{op} is the operation time of the sensor—assumed

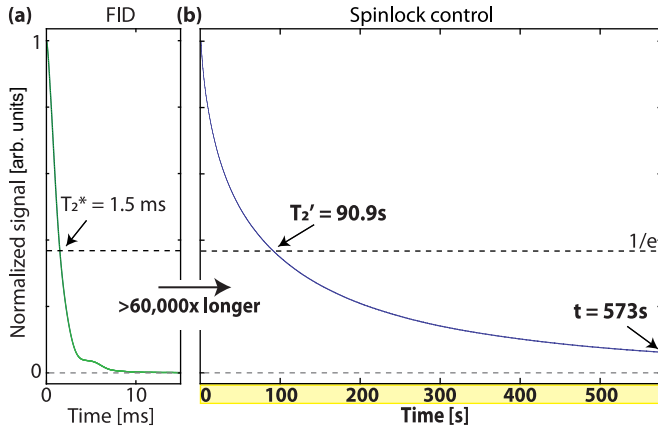


FIG. 6. Effect of spin-locking. (a) Typical ^{13}C FID occurs in ≈ 1.5 ms. (b) Pulsed spin-locking leads to prethermalization, extend the lifetime by $>60\,000$ -fold to $T_2' \approx 91$ s. Spins are continuously interrogated here for 573 s. Data here is reproduced from Ref. [11] for completeness, with the consent of the authors.

to be $T_2^*/2 = 0.75$ ms. The sensitivity is then calculated by normalizing this minimum detectable field by multiplying with the square-root of the total experiment time, including the operation time ($T_2^*/2 = 0.75$ ms) and the dead time for hyperpolarization (40 s). Note that the dead time is much larger than the operation time, however, increasing the dead time tends to also increase the optimal operation time. Using these two assumption we find a sensitivity of $480 \text{ nT}/\sqrt{\text{Hz}}$, as reported in the main text.

Let us emphasize, that in our setup neither of the assumptions are entirely valid since the signal is expected to decay with time so every subsequent shot is actually less sensitive to the field and the decay of the signal seems to follow a stretched exponential profile (see Appendix B 4). However, independent of either of those assumptions, whether the sensor should be operated for one more shot is simply a trade-off between whether that next shot will contribute enough signal to account for the cost of increasing the total experiment time. Given the long dead time, one might thus be tempted to operate the sensor for a time longer than $T_2^*/2$ to increase the sensitivity of the sensor. We can estimate a lower bound on the optimal sensitivity of the Ramsey scheme by allowing for the maximal operation time of $T_{\text{op}} \approx 10$ ms where the signal has all but decayed, see FID in Fig. 6(a), and assuming that the signal does decrease within this time. Using these assumptions the obtained Ramsey sensitivity of $131.5 \text{ nT}/\sqrt{\text{Hz}}$ is still well above the sensitivity of our scheme.

APPENDIX B: DETAILS OF THE EXPERIMENT

1. Sample and hyperpolarization strategy

The sample used in this work is a CVD fabricated single crystal diamond sample that contains ~ 1 ppm NV centers, and natural abundance of ^{13}C concentration. This corresponds to the diamond ^{13}C nuclei being distributed with a spin density $\sim 0.92 \text{ nm}^{-3}$ in the lattice. The sample is placed flat, i.e., with its [100] face pointing parallel to the hyperpolarization and interrogation magnetic fields (38 mT and 7 T, respectively).

Hyperpolarization is carried out at $B_{\text{pol}} = 38$ mT through a method previously described [20,21]. It employs continuous optical pumping and swept microwave irradiation for ~ 40 – 60 s, and ratchetlike polarization transfer develops through the traversal of rotating frame Landau-Zener anti-crossings [68]. Spin diffusion serves to transfer polarization to bulk ^{13}C nuclei in the diamond lattice.

2. Experimental setup

We now provide more details of the apparatus employed for ^{13}C spin control and interrogation in our experiments. In reality, the ^{13}C nuclei are hyperpolarized at room temperature at low-fields (~ 38 mT), and subsequently “shuttled” to high field (7 T) where they are controlled and interrogated. We refer the reader to Ref. [69] for details on the hyperpolarization apparatus and mechanism, and Ref. [70] for details on the shuttling apparatus.

Specifically, the detection of the spins proceeds via inductive coupling to an RF cavity and measurement of corresponding ^{13}C nuclear magnetic resonance (NMR) signals at high field (7 T), where the ^{13}C resonance frequency is 75.38 MHz. This is accomplished via a homemade NMR probe that was detailed in Ref. [50]. It consists of a high RF homogeneity saddle coil that allows > 1 M pulses to be applied to the nuclear spins at high power (~ 30 W) and high duty cycle ($\sim 50\%$). This RF coil is in a saddle geometry and is laser-cut out of OFHC copper with three turns and a coil height of 1 cm. We measure a Q factor of 50 at 75 MHz and a sample filling factor of ~ 0.15 . The probe also has a z coil that is employed to apply the time-varying AC field to the sample. This is a loop of a 2-turn coaxial cable that is wound outside the RF coil (see Ref. [50]).

The AC field itself is produced with a Tabor TE5201 arbitrary waveform generator, and is amplified by an AE Techtron 7224 amplifier into the z coil. The spin-lock RF pulses, however, are produced by a Varian NMR spectrometer. High signal-to-noise detection of the ^{13}C nuclear precession is carried out with a quarter wave line and bandpass filter combination following a transmit/receive switch, and the signal is digitized by a high-speed arbitrary waveform generator (Tabor Proteus). The high data acquisition rate of the device (1 GS/s) allows for high-fidelity sampling of the ^{13}C induction signal between the pulses [50]. The entire experiment is synchronized via a PulseBlaster digital pulse generator (100 MHz).

3. Spin-lock pulse sequence and data processing

For clarity, we describe in Fig. 6 the effect of the spin lock sequence, reproducing data in Ref. [11]. The ^{13}C free induction decay in diamond is $T_2^* \approx 1.5$ ms on account of interspin interactions [Fig. 6(a)]. However, as demonstrated in Fig. 6(b), under pulsed spin-lock it is possible to prolong this coherence to $T_2' > 90$ s. In typical experiments, in this paper, pulse duty cycle is maintained high (19–54%), and interpulse spacing $\tau \sim 100 \mu\text{s}$. Flip angle ϑ can be arbitrarily chosen, except for $\vartheta = \pi$ [31].

Let us now provide more detail about the data processing. For each t_{acq} window between the SL pulses, we directly digitize the heterodyned Larmor precession of the nuclear spins

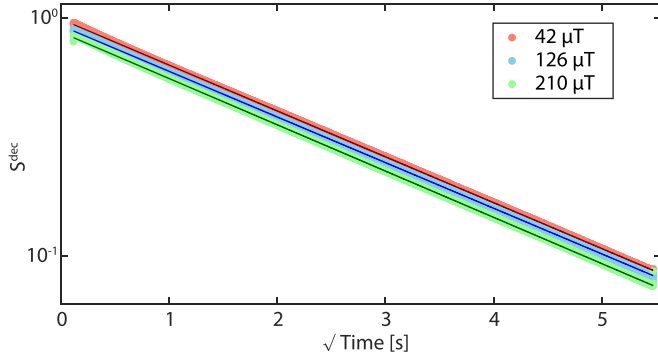


FIG. 7. Stretched exponential nature of S^{dec} . As a complement to the data in Figs. 3(e) and 3(f) of the main paper, we plot the decay profile of signal S in a graph considering S^{dec} vs $\sqrt{t}$ in a semilogarithmic scale for three representative values of B_{AC} . Solid lines are linear fits. Extracted T_2' values for full dataset are plotted in Fig. 3(f).

every 1 ns using a fast AWT (Tabor Proteus). The amplitude and phase of the Fourier transform of the obtained precession [Figs. 9(a) and 9(b)], sampled at $f_{\text{het}} = 20$ MHz (the heterodyned Larmor frequency), then directly provide information of the transverse components of the spin vector [as shown in Fig. 9(c) of the main paper]. This homodyne readout is very effective because of the slow Larmor precession and rapid sampling allows the ability to completely capture the signal with > 100 points per precessing period. This oversampling enables high SNR reconstruction due to the ability to suppress noise at any component except at f_{het} . Ultimately, each readout window provides one such amplitude and phase point, and in typical experiments we apply $N \gtrsim 200$ k pulses.

4. Stretched exponential decays

In this section, we consider the decay profiles of the obtained signal in Fig. 3(e) in the main text and demonstrate that they follow stretched exponential profiles $\propto \exp[-(t/T_2')^{1/2}]$ to a good approximation. To see this, we plot in Fig. 7 the signal S^{dec} for three representative B_{AC} values in a logarithmic scale with respect to $\sqrt{t}$. As in Fig. 3(e), the four manifolds of the raw signal S [see Fig. 2(b)(i)] are smoothed over to produce respective decay profiles S^{dec} . Solid lines are linear fits, and are seen to be linear to a good approximation over the $t = 35$ s period considered. This shows that the decays follow very closely a stretched exponential $\propto \exp[-(t/T_2')^{1/2}]$. We also have previously observed stretched exponential behavior in the prethermalization decay profiles extending to $t > 500$ s [11]. In Fig. 3(f), we extract the respective time constants and calculate error bars from the corresponding data in Fig. 3(e). This same method is employed to report decay constants in Fig. 16(c).

APPENDIX C: DETAILS ON THE OBSERVATION OF PRETHERMAL SPIN TRAJECTORIES

1. Principle of continuous spin trajectory tracking

In this Appendix we describe the principle of spin tracking employed in this paper; schematically shown in Fig. 8(a). Indeed, considering the protocol in Fig. 8(b), the pulsed SL

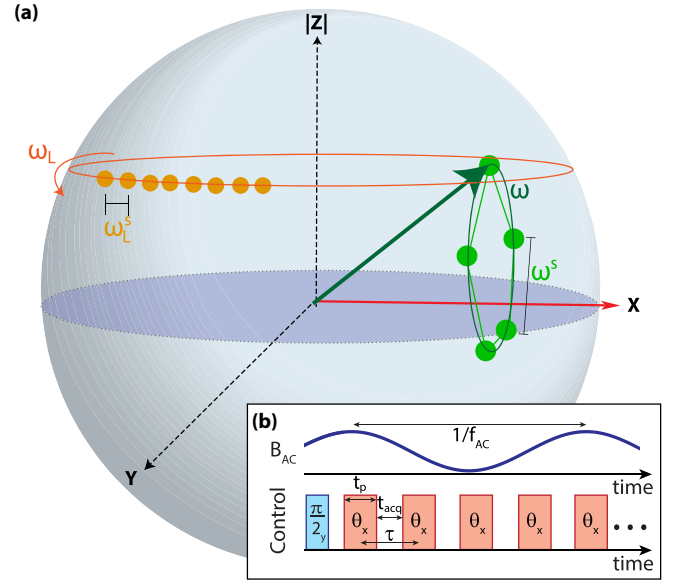


FIG. 8. Spin tracking principle shown for AC field $\mathcal{H}_{\text{AC}}$ applied simultaneously with a spin-locking field along $\hat{x}$ [schematic in panel (b)]. (a) In the rotating frame, spins (green arrow) undergo orbital motion (green circle) under the joint action of both fields; the AC field frequency ω_{AC} is matched to that of a full precession, which completes in time $(\omega/(2\pi))^{-1} = (t_{\text{pulse}}/\tau)\Omega$. Trajectory is sampled at rate ω^s (green points) after each SL pulse; at resonance, $\omega^s = (2\pi/\vartheta)\omega$ leading to the polyhedrally sampled trajectories in Fig. 12 (here a pentagon is sampled). For each such sample, the spins are interrogated by an RF cavity via spin precession in the laboratory frame (orange circle), at rate ω_L . This precession is sampled at rate ω_L^s (orange points). Hierarchical timescale separation (see Table I) ensures the quasicontinuous tracking of the trajectory at ω^s with high SNR.

pulses *dress* the spins such that the energy gap $\omega = \eta\Omega$, where Ω is the Rabi frequency, and $\eta = t_{\text{pulse}}/\tau$ is the pulsing duty cycle. Micromotion under the action of the control field $\mathcal{H}_{\text{AC}}(t)$ results in spin evolution in a rotating (dressed frame) trajectory. In our experiments, this rotating-frame spin trajectory is “sampled” quasicontinuously at a rate $\omega^s/2\pi = \tau^{-1}$ (green points). Practically, this is accomplished by interrogating the spins via an RF cavity by allowing them to precess in the laboratory frame (orange circle) at Larmor frequency ω_L in $t_{\text{acq}}(\sim \tau/2)$ windows between the SL pulses. Crucially, cavity interrogation is arranged to be very rapid: if the rotating-frame trajectory is sampled at rate ω^s , then for each such sample the cavity readout is carried out multiple times, sampled at a rate ω_L^s (orange points) such that $\omega_L^s/\omega^s > 10^4$. Our strategy therefore revolves around ensuring a hierarchal timescale separation (see Table I) such that, $\omega_L^s \gg \omega_L \gg \omega^s > \omega > \gamma B_{\text{AC}}$. This leverages the low ω_L (relative to ω_L^s) Larmor frequencies of the nuclear spins, and their ability to be driven for long periods (T_2'), but interrogated rapidly (at ω_L^s). Table I shows typical values as employed in experiment; the orders of magnitude overall timescale separation proves the key to *quasicontinuous* state tracking.

To see this more clearly, consider some comparisons in Table I. First, the wide separation $\omega_L/\omega \gg 1$, allows the applied AC fields at $\omega_{\text{AC}}(\approx \omega)$ to be filtered from the cavity

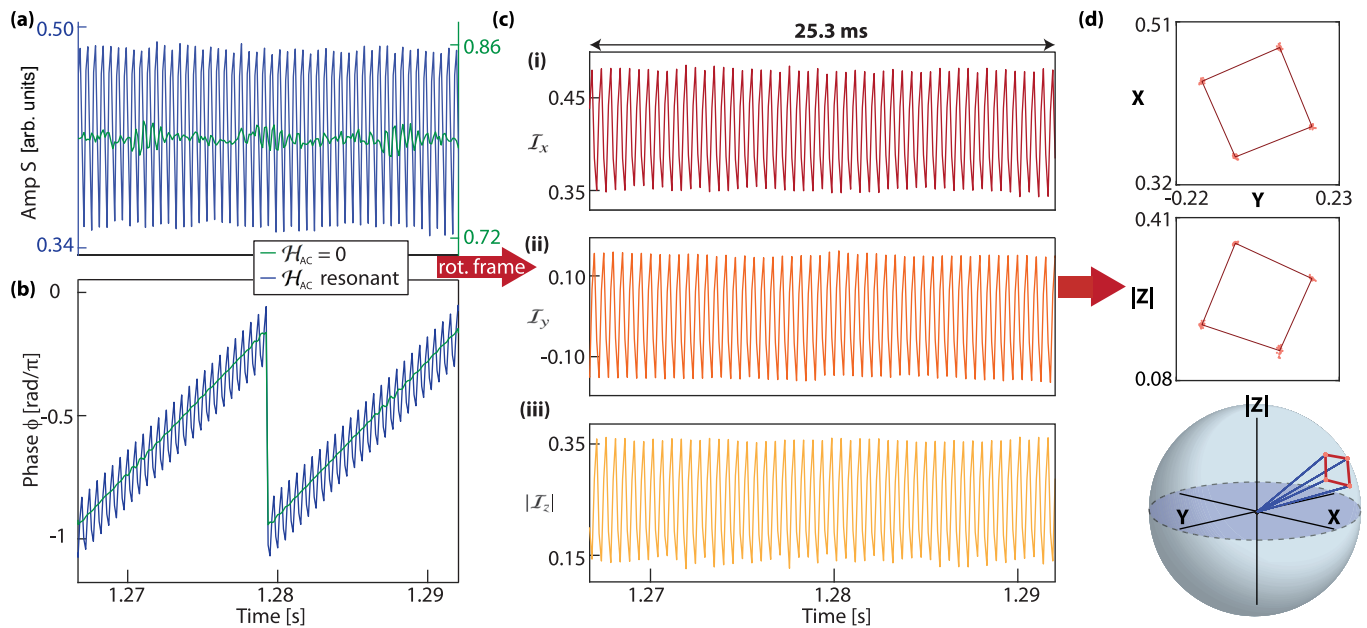


FIG. 9. Principle. (a), (b) *Amplitude and phase response* showing signals S and ϕ , respectively, for a representative $\Delta t = 25.3$ ms window under: (i) no applied control field $\mathcal{H}_{AC}(t) = 0$ (green), and (ii) a resonant sinusoidal AC field at $\omega_{AC} = (2\pi)1953.125$ Hz (blue). Sequence here employs $\vartheta = \pi/2$, $\omega^s = (2\pi) 7.81$ kHz, and full data is collected over 35 s (see Fig. 2). In the former case ($\mathcal{H}_{AC}(t) = 0$), spins are locked along $\hat{x}$. Small wiggles are due to 60 Hz noise pickup (see Appendix B). Ramplike phase pattern indicates phase accrued during pulses. (c) *Dressed frame cartesian trajectory* components. Here (i), (ii) $I_{x,y}$ components are unwrapped from data in panels (a), (b), and (iii) $|I_z|$ is calculated via a unitary constraint. (d) *Bloch sphere representation* of data in panel (c) shown as 2D projections (upper panels), and in three dimensions (lower panel). Points display the sampled rotating frame trajectory over Δt , which manifest in four distinct sets of points. Tracing the centroid of the four sets of points results in a trajectory resembling a quadrilateral.

response (ω_L). This permits RF spin interrogation to proceed simultaneously with the application of the AC field $\mathcal{H}_{AC}(t)$. Second, the hierarchy $\omega^s \ll \omega_L$ means that cavity interrogation can be carried out for multiple Larmor cycles for each sampled point in the rotating-frame spin trajectory [Fig. 8(a)]. This yields a high readout signal-to-noise (SNR) per sampled point on the trajectory, and $\omega^s \gg (T_2')^{-1}$ means that the tracking is for most practical purposes, quasicontinuous. Finally, the large ratio ω_L^s/ω_L means it is possible to carry out exquisite filtration of the cavity response, suppressing all components except those exactly at ω_L , also yielding SNR gains.

2. Three-dimensional spin reconstruction strategy

Following the principle employed in Appendix C 1, let us now describe how the position of the spins is tracked in three-dimensions on a Bloch sphere. Methodologically, we rely on digital homodyne detection of the cavity response at frequency ω_L (Appendix B). In particular, a Fourier transform (FT) of the cavity readout in each SL readout window (separated by τ) is carried out, and the amplitude and phase at ω_L report on the components of the spin vector along the equatorial $\hat{x}$ - $\hat{y}$ plane in Fig. 8(a). For clarity, we will use t_j to denote the quasicontinuous time variable discretized by τ in these measurements. Figures 9(a) and 9(b) then illustrates the data obtained in a 25.3 ms time window with $\vartheta = \pi/2$ for two exemplary cases: (i) first, in the absence of the AC field, i.e., $\mathcal{H}_{AC} = 0$ (green line), i.e., and (ii) second, with an

AC field applied on resonance, $\mathcal{H}_{AC} = B_{AC} \cos(\omega_{res}t)$ (blue line), the case of the “square-trajectory” in Fig. 12(c)(i). When $\mathcal{H}_{AC} = 0$, the constant amplitude signal $S(t_j)$ reflects the spins being locked along $\hat{x}$. However, considering the phase [Fig. 9(b)], ramps arise due to a trivial phase accrual under Larmor precession during each ϑ pulse. Removing this phase via a linear fit (see Appendix B) allows us to extract the effective phase evolution in the dressed frame, $\phi(t_j)$. Combining information from the two quadratures then allows direct access to the transverse projections [see Fig. 9(c)], $I_x(t) = S(t) \cos(\phi(t))$, and $I_y(t) = S(t) \sin(\phi(t))$, where I_v specifies polarization in the rotating frame. I_x and I_y for the square trajectory AC field case are displayed in Figs. 9(c)(i) and 9(c)(ii). The four-point periodic oscillations reflect micromotion driven orbital motion in the rotating frame.

In principle, I_x and I_y is the maximal extent of information that can be extracted from these measurements. However, as is evident in Fig. 9(c), the spins undergo very little decay during successive cavity interrogation windows, $(\omega^s T_2')^{-1} \ll 1$. This allows the ability to extract the full 3D vector $\vec{I}$ information from only homodyne measurements of signal and phase, by reasonably estimating also the $I_z(t_j)$ component (sans its sign) using, $|I_z(t_j)| = [\mathcal{N}^2(t_j) - I_x^2(t_j) - I_y^2(t_j)]^{1/2}$ [Fig. 9(c)(iii)], where norm $\mathcal{N}(t) < 1$ accounts for spin decay up until time t . Following Fig. 3(e), the decay profile $\mathcal{N}(t)$ reflects the prethermalization process. Combining this information to form $\vec{I}(t_j)$ allows us to plot (track) the dressed-frame dynamics on a Bloch hemisphere, shown in Fig. 9(d) for the individual projections on the $\hat{x}$ - $\hat{y}$ and $\hat{y}$ - $\hat{z}$, and in three

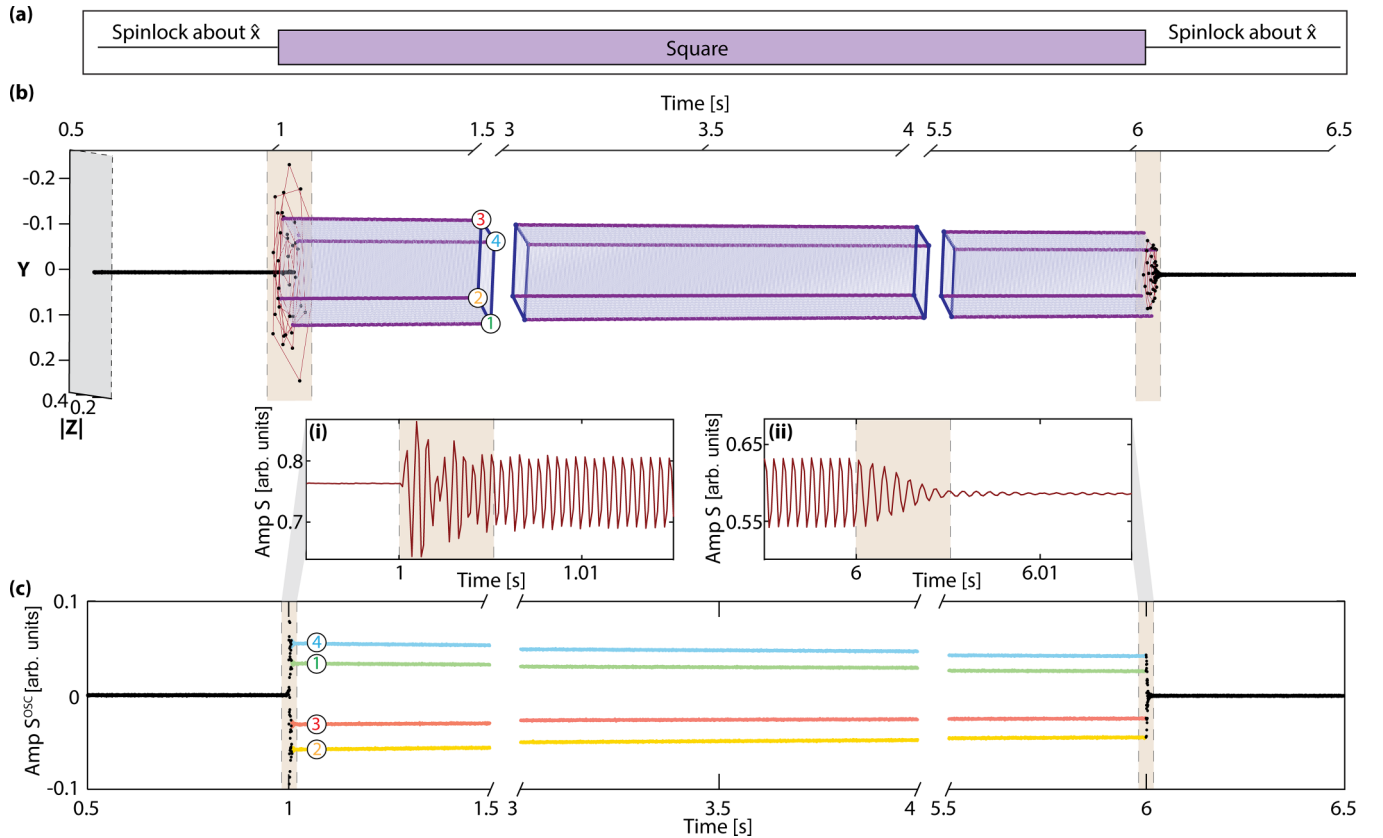


FIG. 10. Emergence of a stabilized squarelike spin trajectory probed via a pulsed AC field. (a) *Schematic*. Spins are subject to pulsed spin-locking along $\hat{x}$ until $t = 1$ s, and then are exposed to an AC field causing a squarelike trajectory for $\Delta t = 5$ s. Subsequently, the field is turned off and the spins rethermalize along $\hat{x}$. Interpulse spacing $\tau = 128 \mu\text{s}$ and $\omega_{\text{AC}} = (2\pi)1953.125 \text{ Hz}$ (see Fig. 12). (b) *Continuously tracked trajectory* shown in the $\hat{y}$ - $\hat{z}$ plane, with a horizontal dimension representing time. Data is averaged over 5 slices everywhere, except in the beige shaded regions (denoted by dashed lines) to discern the transient dynamics (red traces). Broken time axis highlights square cross sections of the spin trajectory [with points labeled 1–4 according to the manifolds in panel (c)]. Slow signal decay is reflected in the decreasing size of the square cross sections over time. (c) *Oscillatory amplitude* S^{osc} as in Fig. 2(c)(i). Splitting of the $\hat{x}$ -locked spins into the four manifolds (circled 1–4) of the square trajectory, their collapse back to $\hat{x}$, and intermittent transient responses (shaded beige regions) are apparent here. Insets (i)–(ii): Zoom into raw amplitude response S at the two transient regions (shaded beige). Transient lifetime is $\sim 4 \text{ ms}$, comparable to native T_2^* .

dimensions on a Bloch sphere. In the 25.3 ms period shown, there are ~ 50 precessions about $\hat{z}$ [see Fig. 9(d)]; the points here display the ω^s samples [Fig. 8(a)] and the lines join their centroids for clarity. This manifests as the quadrilateral spin projections observed in Fig. 9(d) [also highlighted in Fig. 2(d)].

We now elucidate upon the assumptions employed for this reconstruction. The instantaneous norm of the spin vector $\mathcal{N}(t)$ is hard to measure quantitatively because a drop in the signal S can arise both due to relaxation and the tilt of the nuclear spin vector away from the $\hat{x}$ - $\hat{y}$ plane, which are hard to distinguish. Although the prethermal lifetimes are almost identical in the absence and presence of $\mathcal{H}_{\text{AC}}$, there is still slight variation with the magnitude of B_{AC} employed [Fig. 3(f)]. This makes an absolute normalization strategy employing the $\mathcal{H}_{\text{AC}} = 0$ case as a reference difficult. To circumvent this problem, we focus instead on only imposing that the *relative* normalization between two time instants in the spin trajectory are accurate. For this, in the long-time reconstructions in this paper (e.g., in Fig. 2), we assume $\mathcal{N}(t)$ can be evaluated by a small offset $S^{\text{dec}}(t) + \delta_0$, where $\delta_0 =$

$0.05 \times S^{\text{dec}}(t = 0)$. While this yields a (constant) error ($\propto \delta_0^2$) in the absolute values of the projections on $\hat{x}$ axis, it does not affect the relative relationship of spin trajectory projections at different instants t_j of the spin evolution, still yielding a semiquantitative real-time 3D map of the spin trajectories. This is illustrated by comparison of the two panels in Fig. 2(d), where the relaxation-mediated shrinking of the Bloch sphere trajectories are evident at long times.

3. Emergence of stable spin trajectories

Let us now consider the *emergence* of these stable spin trajectories, starting first with the spins locked along $\hat{x}$. Figure 10 shows the evolution of the tracked trajectory upon “turning on” the B_{AC} field corresponding to a “squarelike” trajectory at $t = 1$ s, and subsequently turning it off at $t = 6$ s [see schematic in Fig. 10(a)]. In a visualization focused on the $\hat{y}$ - $\hat{z}$ projection [see Fig. 10(b)], the spins start off tracing a line along the time dimension, subsequently settling into a stable “squarelike” trajectory under B_{AC} (stability evident from the cross sections), that finally collapses back into a line

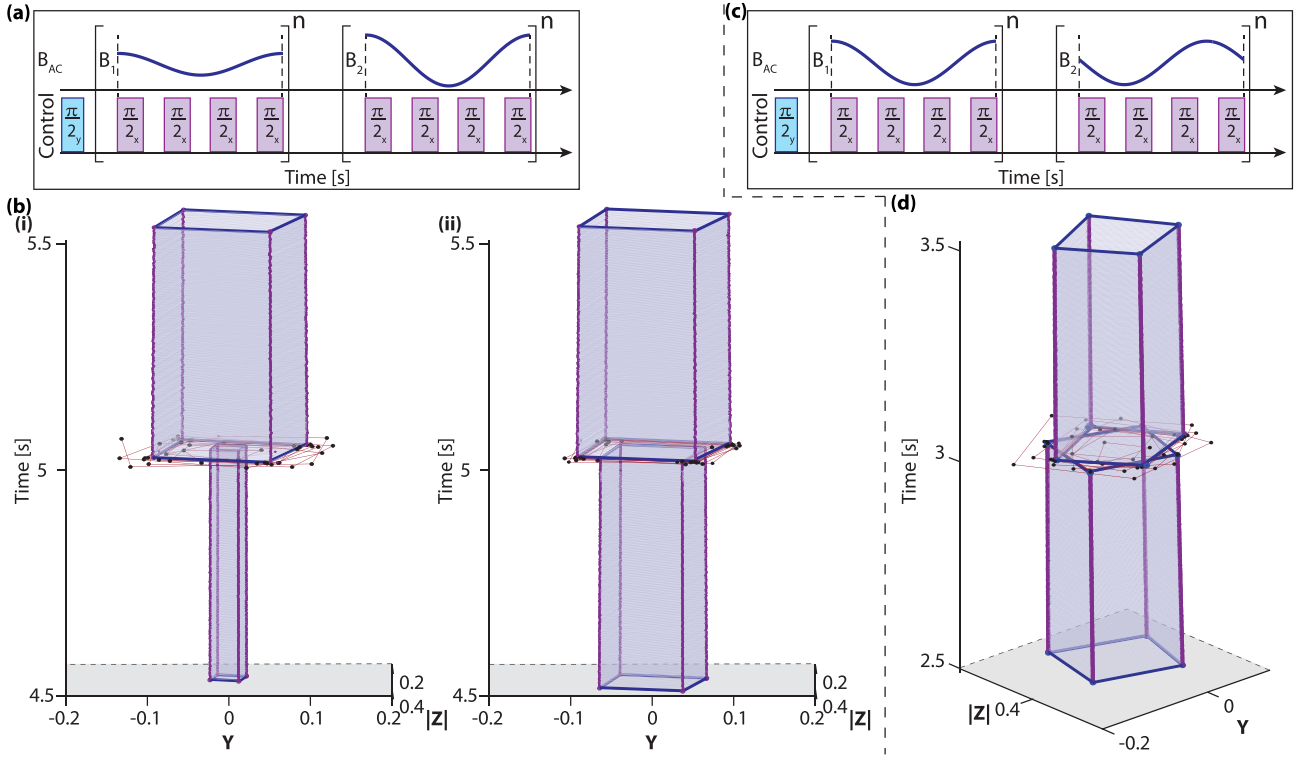


FIG. 11. Transient dynamics upon amplitude and phase quench. (a) *Schematic for amplitude quench* showing control sequence creating a squarelike trajectory under an AC field B_1 for $t = 5$ s (first bracketed portion), which is then switched to an AC field B_2 with higher amplitude for $t = 5$ s (second bracketed portion). (b) (i)–(ii) *Tracked spin trajectories* shown near the switching region for two experiments corresponding to $B_1 \approx \{42, 126\}$ μT and $B_2 = 185$ μT in panel (a), respectively. Emergence of a new stable trajectory is associated with a transient (red traces). (c) *Schematic for phase quench* similar to (a), but with AC field instead phase-shifted by 45° at $t = 3$ s. (d) *Tracked spin trajectories* with $B_1 = B_2 = 185$ μT . Intermittent transient region (red traces) is again visible.

when B_{AC} is turned off. Both switching events are associated with rapid transient dynamics, captured by the red traces in the dashed shaded regions in Fig. 10(b). In this visualization, averaging over 5 frames is carried out everywhere except in the dashed shaded regions to preserve the transients. Figure 10(c) shows the full S^{osc} data with the manifolds labeled following the same convention as in Fig. 2(c)(i) [corresponding edges are marked in Fig. 10(b)].

The transients capture the rethermalization of the spins upon the Hamiltonian quench [71] under the switching action of B_{AC} . To quantify the timescale for this thermalization, insets Figs. 10(c)(i) and 10(c)(ii) show the raw amplitude signals S in the dashed transient regions. We discern a transient lifetime of ≈ 4 ms in both cases, and the close resemblance to T_2^* suggests that thermalization here is driven by interspin interactions.

The transient dynamics are strongly model-dependent and do not exhibit universal features inherent to the subsequent prethermal plateaus. A closed-form analysis is out of reach for our nonintegrable dipolar system; in addition, numerical simulations on a classical computer are challenging. Nevertheless, via an understanding of the thermal properties of the pre- and post-quench equilibrium state, it is possible to make a few statements about the properties of these “on” and “off” processes. In particular, when the AC field is turned on, the system undergoes transient dynamics from the initial state $\tilde{\rho} \propto I_x$ to the prethermal state $\rho_F \propto \beta \mathcal{H}_{F;k}^{(0)}$. In contrast,

turning the AC field off yields the prethermal state $\rho_F \propto \beta \mathcal{H}_{F;k}^{(0)}$ ($B_{AC} = 0$) at the *same* (inverse) temperature β , but with regard to a different Hamiltonian. Note that $\mathcal{H}_{F;k}^{(0)}(B_{AC} = 0)$ is independent of k and differs from $\mathcal{H}_{F;k}^{(0)}(B_{AC} \neq 0)$ by the emergent on-site field, i.e., $\tilde{\mathbf{w}}_k(B_{AC} = 0)$ has only a nonvanishing $\hat{\mathbf{x}}$ component. While it is difficult to make rigorous statements about the transient, these observations suggest that switching the AC field on can lead to a rich transient behavior whereas the “off” transients only correspond to a simple decay of the N plateaus into a single one (see Appendix D4).

In fact, the insets in Fig. 10(c) show clear differences between the two transients. While turning on the AC field appears to cause “spikes” in the signal [Fig. 10(c)(i)] where S momentarily goes higher or lower than the prethermal oscillations, the off-transient is free from large oscillations and only preserves terms in ρ collinear with $\hat{\mathbf{x}}$ which are subsequently stabilized by prethermalization.

To experimentally quantify the size of the transients, in Fig. 11 we consider traced trajectory evolutions under amplitude quenches corresponding to an abrupt change in AC field amplitude [Figs. 11(a) and 11(b)] or phase [Figs. 11(c) and 11(d)]. In particular, we expose the spins to a large ΔB_{AC} from $B_{AC,1} \approx 42$ μT to $B_{AC,2} \approx 185$ μT [Fig. 11(b)(i)] and a smaller ΔB_{AC} from $B_{AC,1} \approx 126$ μT to $B_{AC,2} \approx 185$ μT [Fig. 11(b)(ii)]; additionally, we conduct a phase step change ($\phi_{AC,1} = 0^\circ$ to $\phi_{AC,2} = 45^\circ$) under constant B_{AC} amplitude

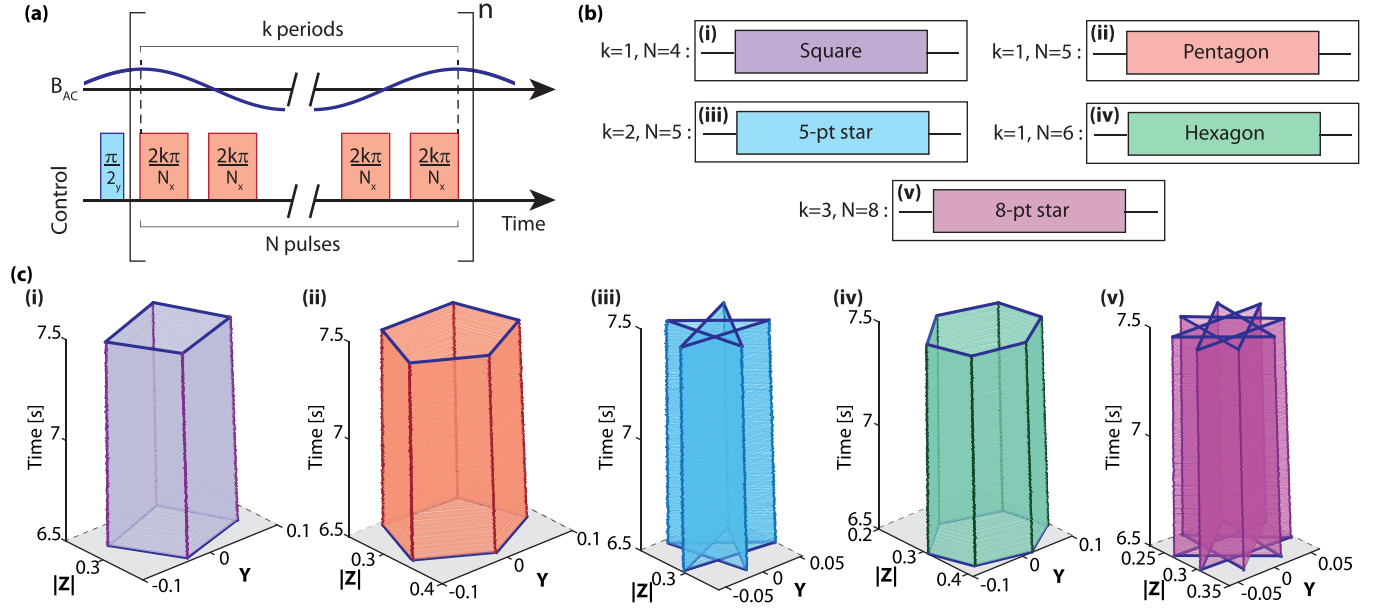


FIG. 12. Long-time engineering and tracking of polyhedral spin trajectories. (a) *Control sequence* consists of a train of pulses of angle $\vartheta = 2k\pi/N$ simultaneous with $\hat{z}$ -axis AC field with frequency such that k periods are matched to N total pulses. Bracketed portion is repeated n ($\gg 1$) times. (b) *Block diagram* of sequences for different polygon spin trajectories (i)–(v), showing corresponding k and N parameter values. (c) *Polyhedral spin trajectories* can be stabilized for multiple second long periods. Shown here are $\Delta t = 1$ s sections of the tracked trajectories in the $\hat{y}$ - $\hat{z}$ plane of the Bloch sphere; time is on the vertical axis. Top and bottom faces are highlighted in blue for clarity. For the different polyhedra [(i) square, (ii) pentagon, (iii) five-point star, (iv) hexagon, and (v) eight-point star], $\omega_{AC} =$ (i) $(2\pi) 1953.125$ Hz, (ii) $(2\pi) 1801.8018$ Hz, (iii) $(2\pi) 1117.3184$ Hz, (iv) $(2\pi) 1673.3601$ Hz, and (v) $(2\pi) 733.1378$ Hz, respectively (see Appendix C 4 and Ref. [72] for movies of motion). Measured lifetimes $T_2' =$ (i) 28.0 ± 0.3 s, (ii) 27.8 ± 0.2 s, (iii) 28.1 ± 0.2 s, (iv) 25.7 ± 0.3 s, (v) 26.4 ± 0.1 s. We average over every two slices along the time axis for clarity; plots shown correspond to an average over 30 experiments.

[Fig. 11(d)]. The corresponding control sequences are described in Figs. 11(a) and 11(c). Appendix D 4 provides a more detailed theoretical discussion of the quench dynamics and associated transients.

4. Polyhedral trajectory shapes

Figure 12(c)(i) shows the tracked motion in Fig. 2(c) for a $\Delta t = 1$ s window centered at $t = 7$ s. The spin evolution traces an approximately rhombohedral prism (here the top and bottom faces are highlighted in blue for clarity). Stability of the motion is particularly clear in the straightness of the prism sidewalls. The rotational orientation of the prism depends directly on the AC phase ϕ_{AC} in Eq. (3) (for Fig. 12(c)(i), $\phi_{AC} = 0$). Data plotted here comprises 7813 pulses (separated by $\tau = 128$ μ s), and for clarity we average over 3 consecutive points in each manifold.

Spin stabilization and tracking can be extended to other shapes via the generalized protocol described in Fig. 12(a). It comprises $\vartheta = 2\pi k/N$ pulses and imposes that ω_{AC} is such that the AC field completes k full periods every N pulses, i.e., $\omega_{AC} = 2\pi k/(N\tau)$. We will refer to the AC field in this case as satisfying a *resonance condition*, with ω_{AC} here being the “resonant frequency” $\omega_{AC} = \omega_{res}$. In reality, as detailed in Appendix C 5 b, this condition can be even more relaxed and made robust against error in ϑ .

Sequences in Fig. 12(b) then produce different “polyhedral” spin trajectories, the tracked results of which are shown in Fig. 12(c) for a (i) square, (ii) pentagon, (iii) five-point star,

(iv) hexagon, and (v) eight-point star. The latter is also shown on a Bloch hemisphere in Fig. 1(c). Measured T_2' lifetime values of the corresponding trajectories are listed in caption of Fig. 12(c). The five-point and eight-point star [Figs. 12(b)(iii) and 12(b)(v)] represent examples of a family of nonconvex shapes created by employing $\vartheta > \pi/2$ in Fig. 1(b). We note that in Fig. 12(b)(v), a single Floquet period occupies $N\tau = 1.364$ ms, a significant proportion of the coupling induced $T_2^* \approx 1.5$ ms. It is interesting that stabilization of the spin trajectory can be retained even in this limit.

Remarkably, the protocol in Fig. 12(a) will robustly produce polyhedral trajectories even if the AC field applied is significantly distorted from a pure sinusoid as long as the resonance condition is maintained (see Appendix C 5 a for a detailed discussion). This is because the micromotion imposes that each manifold in S^{osc} (corresponding to each edge of the prism) depends, via Eq. (6), on the *full* AC field over one period rather than its instantaneous value after every pulse (see Appendix C 5 a).

Movies and projections corresponding to polyhedral trajectories

As a complement to Fig. 12(c), Fig. 13 presents all three projections of the excited spin trajectories, and compares the motion at $t = 5$ s and $t = 15$ s similar to Fig. 2(d). The panels once again emphasize that the orbital motion is highly stable. A special feature of the visualization here is that the $\hat{x}$ - $\hat{z}$ projections of the motion can be seen to be arclike. This illustrates that the motions in Fig. 12 are akin to 2D polygonal

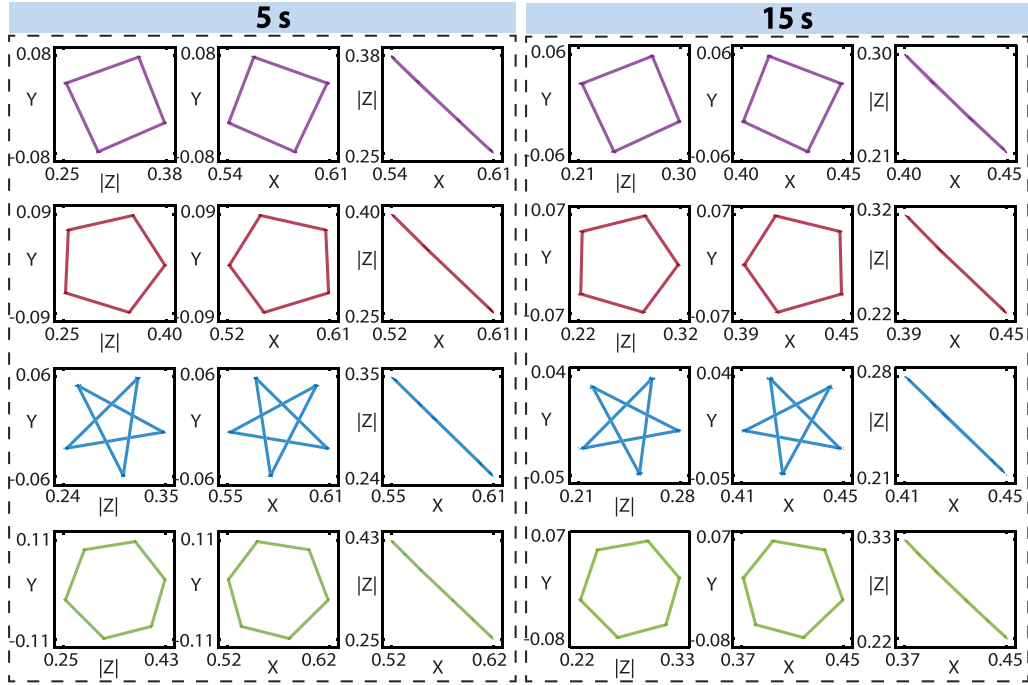


FIG. 13. Bloch sphere trajectories for representative tracked polyhedral spin orbits in Fig. 12(c) shown as individual projections on the $\hat{y}$ - $\hat{z}$, $\hat{x}$ - $\hat{y}$, and $\hat{x}$ - $\hat{z}$ planes. We focus on two $\Delta t = 40$ ms windows of data starting at $t = 5$ s and $t = 15$ s respectively. Data here is shown after 15 averages. Points show the reconstructed spin positions on the Bloch sphere while solid lines join their respective centroid. Stability of the motion is reflected in the relatively small decay between the panels at different time points. Arclike projections in the $\hat{x}$ - $\hat{z}$ plane indicate that trajectories are akin to “tilted” 2D shapes plastered on the 3D Bloch sphere. Data here is shown as a movie in the Supplemental Material [72].

shapes pasted atop the 3D Bloch sphere. Movies of the tracked motion can be seen in the Supplemental Material [72].

$B_{AC}(t)$ only affects the area enclosed, but not the orbital shape itself (see also Appendix C 5 b).

5. Robustness and stability

a. Robustness of trajectories to amplitude distortion

A remarkable feature of the spin orbits is that they are robust to the exact functional form of the AC field. Indeed, it is possible to obtain orbits similar to Fig. 12(c) even when the AC field is significantly distorted from a sinusoid. An immediate consequence of this resilience is that amplitude fluctuations (noise) in the AC field have very little effect to the obtained spin orbits.

To illustrate this, in Fig. 14 we carry out experiments with an exemplary AC field that is constructed by combining two half-period sinusoids with a five-fold amplitude ratio [schematically shown in Fig. 14(a)]. $\vartheta = \pi/2$ SL pulses are employed here, and the overall period of the AC field is resonant: one full AC field period matches that of four SL pulses [$\omega_{AC} = 2\pi/(4\tau)$]. We then carry out spin tracking on the resulting orbits; as the projections in Fig. 14(b) indicate, the obtained trajectories continue to closely resemble a “square,” as expected for a pure undistorted sinusoid as in Fig. 12(c)(i). The obtained trajectory also remains equivalently rigid, as evident in tracked data in Fig. 14(c). The origin of this robustness can be seen from Eq. (6) where the vector family $\tilde{w}_k$ is constructed out of functions $f_n(\phi_{AC})$ that are evaluated as an integral over the *entire* period T_{period} of the AC field, and not on the instantaneous value of the amplitude $B_{AC}(t)$ in-between the pulses (see also Appendix D 2). Hence, a distortion in

b. Robustness of trajectories to SL flip angle error

In this Appendix, we demonstrate that the “resonance condition” as employed in the main paper can be very relaxed, offering a high degree of robustness with respect to the choice of the SL flip angle ϑ . Indeed, the ability to apply $>250k$ pulses in the presence of RF inhomogeneity means that the obtained stable orbits are highly robust to ϑ . In this section, we consider experimentally the situation when $\vartheta \neq 2\pi/N$, but deviates slightly from it, while remaining the same for all applied pulses.

Figure 15 shows experiments for three representative values of $\vartheta = \{107^\circ, 124^\circ, 130^\circ\}$. In each case we satisfy a *relaxed* version of the resonance condition, only requiring that one AC period completes in a total period $N\tau$, i.e., $\omega_{AC} = \omega_{\text{res}} = 2\pi/(N\tau)$. Experimentally, this condition is easier to satisfy because it only requires a precise period matching between the AC field and the SL pulses.

In Fig. 15, we demonstrate this for a representative case of $N = 4$ and use the notation employed in Fig. 2, showing signal S^{osc} and phase ϕ over a 10s period. The four plateaus demonstrate that the stable squarelike trajectory forms in this case, even though the pulses deviate from $\pi/2$. Overall, Figs. 14 and 15 demonstrate that stable spin orbits are robust against error in both the AC field amplitude and SL pulses. As long as the SL pulses and the AC field are period-matched (mutually resonant), the spin orbits will remain identical and stable.

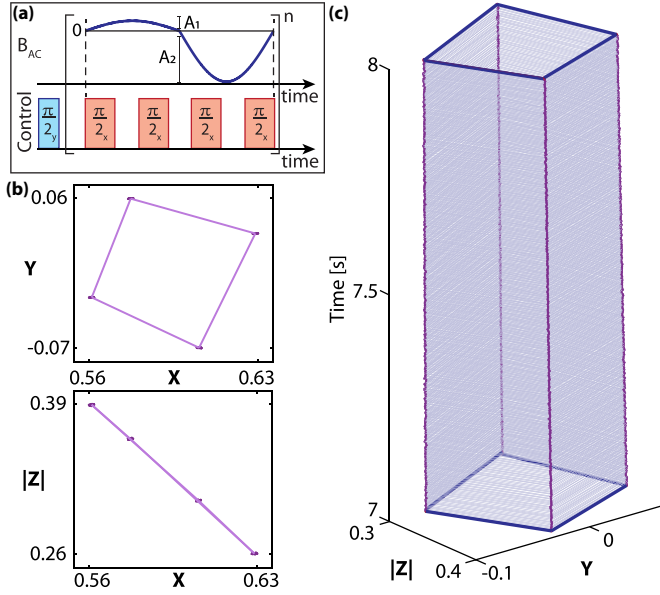


FIG. 14. Robustness of spin trajectory to variations in AC field. Tracked spin trajectory for a representative “distorted” sine-wave AC field. (a) Schematic of experiment; $\vartheta = \pi/2$ and the AC field is arranged to be resonant. Here, the second half-period has amplitude $A_2 = 5A_1$. (b), (c) Tracked spin trajectory in a $\hat{y}$ - $\hat{z}$ plane over time on the vertical axis (c), and as individual $\hat{x}$ - $\hat{y}$ and $\hat{x}$ - $\hat{z}$ projections (b) at $t = 5$ s. Trajectory traced remains “squarelike” in spite of the introduced distortion.

We envision this feature will have important implications for quantum sensing with hyperpolarized nuclear spins.

c. Stability of trajectories

We give further evidence for the robustness of the engineered trajectories, complementing the findings in Sec. III D. For instance in Fig. 4(c) with multiple “opening” and “closing” events of polyhedral spin excursions, are inherently robust. To see this, Fig. 16 shows a comparison between the decay of the trajectory in Fig. 4(c), tracked for $t = 15$ s, to that of a single square trajectory over the same period [see schematic in Fig. 16(a)]. The trajectory here consists of pentagonal and square excursions created with $\omega_{AC} = (2\pi)1801.8018$ Hz and $\omega_{AC} = (2\pi)1953.125$ Hz respectively, which are switched on for $\Delta t = 2$ s in an alternating fashion. Plotted is the full raw signal S ; the rapid AC field driven oscillations appear here as apparent bands in the signal as shown in Fig. 16(b).

It is evident that every switching event [(a)–(d)] in Fig. 16(b) yields a transient. The “closing” transient results in pre-thermalization of the spins so that only the component collinear with $\hat{x}$ is retained [see also Fig. 10(c)(ii)]. Since the switching instants are not exactly matched to a complete period to either ω_{AC} , this leads to a steplike signal jump at every closure. Similarly, an opening transient results in prethermalization to the corresponding $\tilde{w}_k$ trajectory in Eq. (6). Importantly however, as Fig. 16(b) illustrates, the overall decay profile during the excursion periods closely matches that of the original pentagonal one, as can be seen by comparing the envelope of the respective decays in Fig. 16(b)

(S^{dec}). In Fig. 16(c) we plot the smoothed version of the signal in Fig. 16(b), where the individual decays follow stretched exponentials with $T_2' = 32.6$ s and $T_2' = 25.3$ s respectively. Overall, spin trajectories are highly robust even when subject to multiple AC field switching events.

d. Effect of adiabatically varying fields

Frequency offset. Rigidity of the trajectories is also retained when the frequency ω_{AC} deviates from the resonance condition. This is illustrated in Fig. 18 comparing the trajectories on-resonance ($\omega_{AC} = \omega_{\text{res}}$) and off-resonance ($\omega_{AC} = \omega_{\text{res}} + (2\pi)10$ Hz). A “screwlike” trajectory with a 10 Hz pitch (corresponding to $|\omega_{AC} - \omega_{\text{res}}|$) is then traced in Fig. 18(b), with a handedness determined by the sign of the off-resonance detuning.

Frequency chirp. While so far we have considered trajectories under quenched fields, its scope can also be expanded to adiabatically varying fields. As an example, in Fig. 17 we consider experiments under a frequency chirped field $\mathcal{H}_{AC}(t) = B_{AC} \cos(\omega_{AC}(t)t)\hat{z}$, where $\omega_{AC}(t) = \omega_{AC}(0) + \delta\omega t/T$ is a linearly ramped frequency, with $\omega_{AC}(0) = 1$ kHz, $\delta\omega = 2$ kHz, and $T = 35$ s [Fig. 17(a)(i)]. Its effect is to drive a rapid adiabatic passage [73] (RAP) of the spins prepared along $\hat{x}$ toward $-\hat{x}$. This is borne out by the measurements in Fig. 17(a), where we display the net amplitude signal $S(t)$. The sharp dip in the signal corresponds to the spins precessing close to the $\hat{y}$ - $\hat{z}$ plane, and occurs when the instantaneous frequency crosses the dressed frame resonance $\omega_{AC}(t) \approx (t_{\text{pulse}}/T)\Omega$. Figure 17(b) shows the tracked motion on the $\hat{x}$ - $\hat{z}$ projection for chosen instants along the motion (see colorbar). Phase unwrapping of φ becomes more challenging around the resonance condition (see discussion in Appendix B). Figures 17(c) and 17(d) shows the extracted Bloch reconstructions of the spins at two instants ($t = 22$ s and $t = 25$ s), on either side of the RAP resonance condition, clearly demonstrating the adiabatic flipping of the spins.

APPENDIX D: THEORY OF PRETHERMAL SPIN TRAJECTORIES

1. Theoretical model and rotating frame Hamiltonian

In this Appendix, we present a derivation of the rotating frame Hamiltonian in Eq. (1). Consider first that the laboratory frame Hamiltonian of the interacting system of ^{13}C nuclear spins is given by

$$\mathcal{H}_{\text{lab}}(t) = \omega_L I_z + \mathcal{H}_{\text{dd}} + \mathcal{H}_z + \mathcal{H}_{AC}(t) + \mathcal{H}_{\text{SL,lab}}(t), \quad (\text{D1})$$

with the Larmor frequency $\omega_L = \gamma_n B_0$, where γ_n is the magnetogyric ratio and $B_0 = 7$ T is the bias magnetic field. Eq. (D1) also includes the dipole-dipole interaction $\mathcal{H}_{\text{dd}}$, the time-fluctuating on-site fields $\mathcal{H}_z$, and the AC drive $\mathcal{H}_{AC}$, as defined in Eq. (2). Now, in the laboratory frame, the pulsed spin-locking (SL) drive can be described by the Hamiltonian,

$$\mathcal{H}_{\text{SL,lab}}(t) = \cos(\omega_{\text{SL}} t) \Omega \Theta(t) I_x, \quad (\text{D2})$$

where we assume the carrier frequency is applied close to the Larmor frequency resonance, $\omega_{\text{SL}} = \omega_L + \delta$, with the small detuning given by δ (with $\delta/\omega_L \ll 1$).

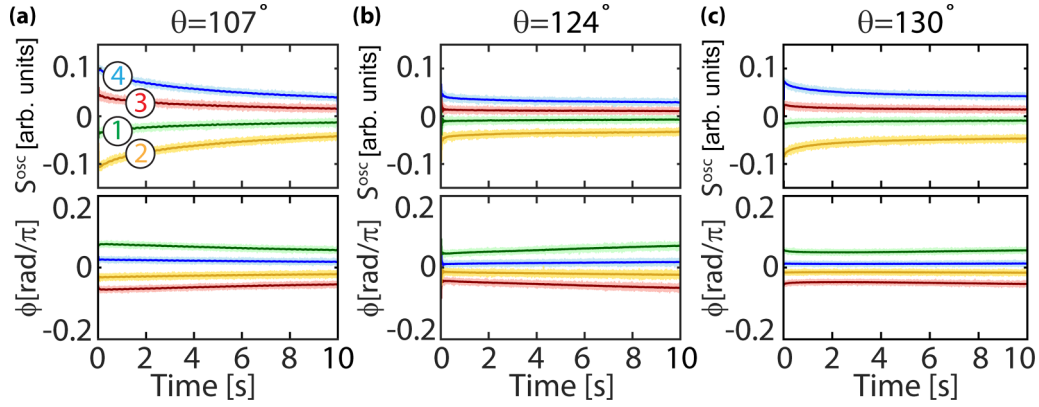


FIG. 15. Robustness to deviations in SL flip angle. Shown are characteristic plateaus shown for signal S^{osc} and phase ϕ similar to Fig. 2(c) of the main paper, but for SL pulse angles ϑ chosen to be $\vartheta = \{107^\circ, 124^\circ, 130^\circ\}$ respectively. AC field frequency is chosen to be resonant in each case, $\omega_{\text{AC}} = \omega_{\text{res}}$ such that one AC period completes in exactly 4τ . Four parallel plateaus (labeled circled 1–4 as in Fig. 2) are visible indicating stable squarelike spin orbits in spite of the ϑ deviating from $\pi/2$.

As is typical at high field, the Larmor frequency ω_L exceeds all other energy scales of the Hamiltonian Eq. (D1) by at least three orders of magnitude (see Table I). We can remove this large energy scale by changing to a rotating frame with respect to $W(t) = \exp(-i(\omega_{\text{SL}}t + \phi)I_z)$. In principle, a generic transformation to the rotating frame can also have a static (i.e., change-of-basis) contribution, which introduces an offset in the experimentally observed phase ϕ [for more details see Appendix D 1]; for simplicity, we will choose $\phi = 0$ hereinafter. Note that the transformation $W(t)$ also affects $\mathcal{H}_{\text{SL, lab}}$ which is transformed to $\mathcal{H}_{\text{SL}} = \Omega\Theta(t)I_x$ upon performing a rotating wave approximation. Taking all these

considerations into account, it is straightforward to arrive at the rotating frame Hamiltonian as specified in Eq. (1),

$$\mathcal{H}_{\text{rot}}(t) = \mathcal{H}_z + \mathcal{H}_{\text{dd}} + \delta I_z + \mathcal{H}_{\text{SL}}(t) + \mathcal{H}_{\text{AC}}(t).$$

We now specify the assumptions we will employ to aid the theoretical analysis. First, the on-site random fields $\mathcal{H}_z$ play an insignificant role for the dynamics of the system, and as such we will ignore them. Second, while the detuning δ is small compared to the Larmor frequency ω_L , we emphasize that it can be comparable to, or larger than, the remaining energy scales in the Hamiltonian, i.e., our experiments can be carried out even in the regime where $\delta \gtrsim J$. In fact, a finite detuning is crucial to theoretically capture the experimentally observed tilt of the spin orbits away from the $\hat{x}$ axis and toward the $\hat{y}$ - $\hat{z}$ plane; see Appendix D 2. Finally, an important hierarchy of energy scales in the rotating frame Hamiltonian Eq. (1) is that the SL Rabi frequency Ω exceeds all other scales by at least an order of magnitude: $\Omega \gg J \approx |\mathcal{H}_{\text{AC}}|$. Therefore, it is sufficient to keep only the contribution of the $\hat{x}$ kicks and detuning δ during the SL pulses. These approximations lead directly to the Hamiltonian in Eq. (3).

Role of the phase in the transformation to the rotating Frame

In Appendix D 1 we introduced the rotation to the dressed rotating frame, $W(t, \chi) = \exp[-i(\omega_{\text{SL}}t + \chi)I_z]$, which features the phase χ . A finite phase, $\chi \neq 0$, corresponds to a constant shift in the experimentally observed dressed frame phase $\varphi = \arctan(I_x/I_y) - \chi$. While the derivations in Appendix D 1 and following are done for $\chi = 0$, one can simply account for the additional phase χ by performing a subsequent static unitary transformation: $W_{\text{static}} = \exp(-i\chi I_z)$ on the final results.

Note that the phase χ naturally enters the expression for the experimentally measured phase φ . However, due to the subtraction of the ramplike profile of the phase φ from the raw experimental data, see Fig. 9 and Appendix C 2, the actual observable becomes $\varphi_0 = \varphi(B_{\text{AC}}) - \varphi(B_{\text{AC}} = 0)$. Therefore, this subtraction procedure rather can be naturally viewed as fixing the gauge freedom associated with the phase χ , so that $\varphi = 0$ at $B_{\text{AC}} = 0$.

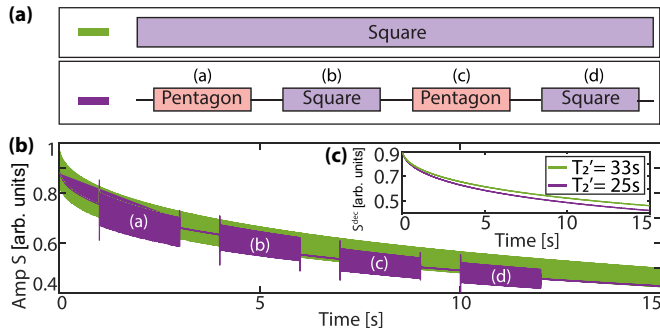


FIG. 16. Decay of designer spin trajectory. Comparison of decay of Fig. 4(c), consisting of multiple separated pentagons and squares, to that of a single square of the same B_{AC} amplitude. (a) Schematic of experiments being compared [cf. Fig. 12(c)(i) and Fig. 4(c)]. (b) Raw signal S plotted in both cases. Motion under B_{AC} appear as bands because the oscillations [see Fig. 2(a)] are rapid compared to the total 15s period under study. Envelope of oscillations allows a means to compare both cases, and demonstrate that the signal decay in each B_{AC} window closely matches that of the original square trajectory. Transient upon each Hamiltonian quench event is clearly visible. (c) Comparison of decay profiles S^{dec} in both cases, obtained by smoothing respective data in panel (b). We measure a relative stretched exponential decay constant of $T_2' = 32.6 \pm 0.3$ s and $T_2' = 25.3 \pm 0.1$ s for the trajectories, demonstrating that the decays remain relatively unaffected even after multiple B_{AC} switching events.

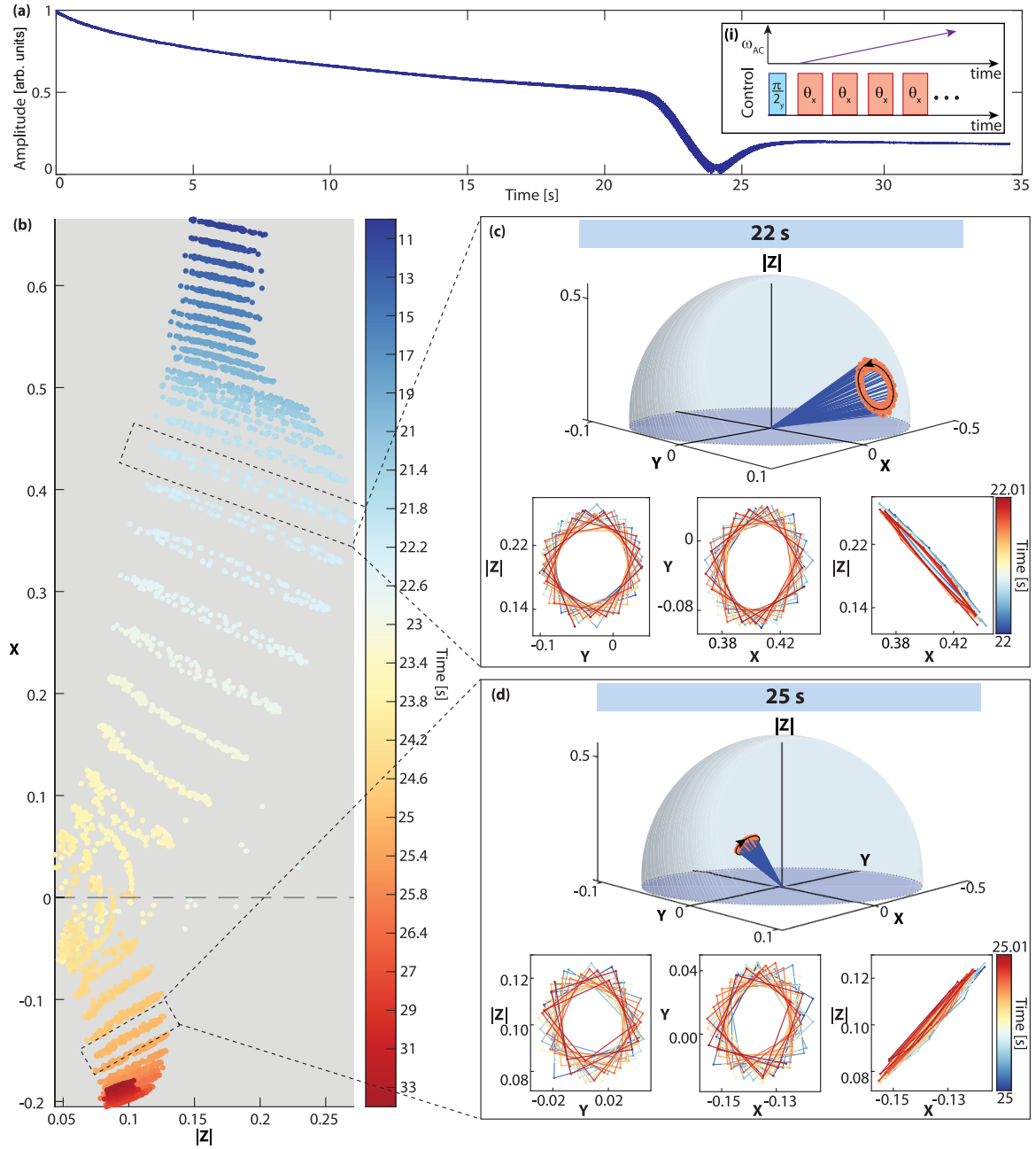


FIG. 17. Continuous spin tracking under rapid adiabatic passage. (a) *Signal amplitude* S under a slow ω_{AC} drive swept in frequency, crossing resonance ω_{res} at $t \approx 22$ s. (i) *Pulse sequence* schematic showing simultaneous control sequence (here $\vartheta = \pi/2$) and AC field with linearly swept frequency ω_{AC} . (b), (d) Tracked spin evolution in three dimensions. (b) Spin evolution in the $\hat{x}$ - $\hat{z}$ plane for various $t = 10$ ms windows, wherein motion resembles closed arcs. Here individual slices are nonlinearly chosen for clarity (see colorbar). (c), (d) Panels display two representative slices at $t = 22$ s and $t = 25$ s, and show full motion for a $\Delta t = 10$ ms interval (colorbar) in a 3D Bloch sphere representation and three 2D projections. Arrows denote the direction of spirallike motion. Spin flip $\hat{x} \rightarrow -\hat{x}$ is evident as a result of the adiabatic rapid passage.

2. Derivation of the leading-order Floquet Hamiltonian

In the following, we derive the family of Floquet Hamiltonians $\mathcal{H}_{F,k}^{(0)}$, given in Eq. (5) of the main text, starting from the simplified Hamiltonian $\mathcal{H}_{rot}(t)$ in Eq. (3) and employing the assumptions elucidated above (see Appendix D 1).

Our discussion will be based on the sequence in Fig. 19(a) for clarity. In general, there exist N distinct unitaries $U_{acq,k+j}$ (labeled by index $k, k = 1, \dots, N$) corresponding to

the different instants $t_k = T_{AC}k/N$ that we consider to represent the start of a period. The Floquet unitary describing the time evolution over one AC period $T_{AC} = 2\pi/\omega_{AC}$ can be written as

$$\begin{aligned}
 U_{F,k} &= U_{acq,k+N} U_{SL} \dots U_{acq,k+2} U_{SL} U_{acq,k+1} U_{SL} \\
 &\equiv \prod_{j=1}^N U_{acq,k+j} U_{SL},
 \end{aligned} \tag{D3}$$

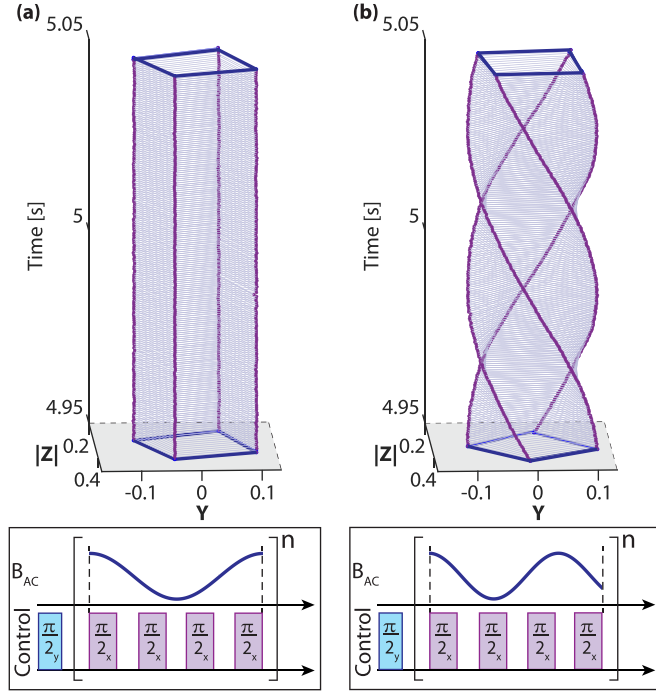


FIG. 18. Spin trajectory response to frequency offset. Tracked spin trajectories for $\vartheta = \pi/2$ in Fig. 1(b), in the case of (a) a resonant AC field ($\omega_{AC} = \omega_{res} = (2\pi)1953.125$ Hz) and (b) an off-resonant field ($\omega_{AC} = \omega_{res} + (2\pi)10$ Hz). Data for a $\Delta t = 0.1$ s window is shown here. *Lower panels:* schematic of the experiments. Degree of off-resonance is exaggerated here for clarity. Frequency offset in panel (b) results in a stable “screwlike” trajectory with a square cross section. Screw pitch is given by the alias frequency ($\omega_{AC} - \omega_{res}$) with handedness determined by the sign of off-resonance deviation.

where $U_{acq,j} = \exp(-it_{acq}\mathcal{H}_{acq})$ and $\mathcal{H}_{acq} = \mathcal{H}_{dd} + \delta I_z + f_j(\phi_{AC})I_z$; $f_n(\phi_{AC}) = \int_{t_n+t_{pulse}}^{t_{n+1}} \sin(\frac{2\pi t}{T_{AC}} + \phi_{AC})dt/t_{acq}$ is the effective phase developed due to evolution between two successive SL pulses. This is highlighted in Fig. 19. The unitary operator modeling the action of the SL pulses can be written as

$$U_{SL} = \exp(-it_{pulse}[\Omega I_x + \delta I_z]) = \exp(-i\vartheta[\cos(\alpha)I_x + \sin(\alpha)I_z]), \quad (D4)$$

where we identify $\vartheta = t_{pulse}\sqrt{\Omega^2 + \delta^2}$ as the total angle acquired during a single pulse, and $\alpha = \arctan(\delta/\Omega)$ represents the deviation of the pulse direction from the $\hat{x}$ axis.

To simplify the analysis, we consider a static change of basis which maps $(\cos(\alpha)I_x + \sin(\alpha)I_z) \rightarrow I_x$. In effect, this allows us to ignore the trivial tilt of the spin orbits away from the $\hat{x}$ axis due to the SL pulse detuning, and introduce it back later. The basis change here amounts to a global static rotation about the $\hat{y}$ axis by an angle $\alpha = \arctan(\delta/\Omega)$, and can be described by the unitary $V = \exp(-i\alpha I_y)$. In what follows, we will denote operators transformed in this basis by a tilde.

We will also consider the experimentally relevant scenario (see Fig. 12) where each of the SL pulses accumulates a flip angle $\vartheta = 2\pi/N + \delta\vartheta$, with $\delta\vartheta \ll 1$ (see also Appendix E2), such that their combined rotation adds up to (almost) unity over one full AC period. To account for the

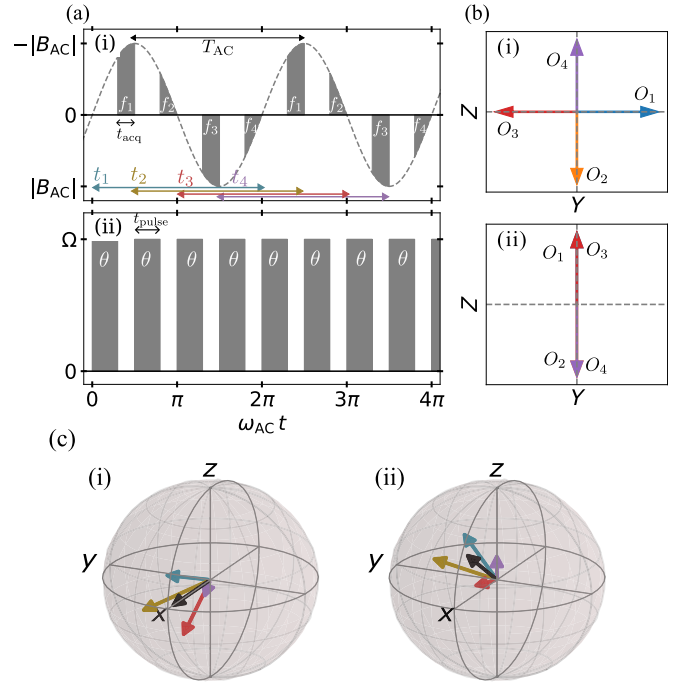


FIG. 19. Theoretical evaluation of spin driving protocol. (a) *Schematic.* Panels show the AC field and SL pulses [see Fig. 1(b)]. For simplicity here $N = 4$ SL pulses are matched to one AC period $T_{AC} = 2\pi/\omega_{AC}$. Shaded regions respectively denote time regions where AC and kicks dominate. Functions f_n , encapsulating the effect of the AC field during the t_{acq} windows, are shown. Families k correspond to evaluating the action of the sequence considering different starting points, $t_k = T_{AC}k/N$, schematically represented by the colored arrows. (b) Schematic representation of operators $O_n = U_{SL}^{-n}I_zU_{SL}^n$ for pulses U_{SL} throughout the sequence, shown for the case of (i) $\vartheta = \pi/2$ and (ii) $\vartheta = \pi$. (c) *Bloch sphere representation* of corresponding polarizations $\mathbf{M}$, plotted for (i) vanishing detuning ($\delta = 0$), and (ii) with finite detuning δ . Colored vectors correspond to case $\vartheta = \pi/2$, whereas the black vector corresponds to the case $\vartheta = \pi$. In the latter case, all four plateaus have equal polarizations. Detuning serves to tilt the orbits away from the $\hat{x}$ axis.

deviation away from $2\pi/N$, we split up the action of the SL pulse in two parts, $\tilde{U}_{SL} = \exp(-i\delta\vartheta I_x)\exp(-i2\pi/N I_x)$. We include the first part into $\tilde{U}_{acq,k}$, i.e., $\tilde{U}_{acq,k} \rightarrow \tilde{U}'_{acq,k} = \tilde{U}_{acq,k}\exp(-i\delta\vartheta I_x)$, and isolate the commensurate kicks $\tilde{U}_{SL} \rightarrow \tilde{U}'_{SL} = \exp(-i2\pi/N I_x)$. For readability we will drop the prime superscript on the unitaries.

To describe the time evolution over stroboscopic times $nT_{AC} + t_k$, where t_k is fixed, we make use of Floquet’s theorem; it states that the stroboscopic evolution is generated by a time-independent Floquet Hamiltonian $\mathcal{H}_{F;k}$ defined via

$$U_{F;k} = \exp(-iT_{AC}\mathcal{H}_{F;k}), \quad (D5)$$

where the index k explicitly indicates that the Floquet Hamiltonian depends on the time instant that defines the start of the sequence, $t_k = k/NT_{AC}$. However, as our model in Eq. (3) is both nonintegrable and long-range interacting, solving for the exact Floquet Hamiltonian is intractable. Instead, we make use of the high-frequency expansion $\mathcal{H}_{F;k} = \sum_n \omega_{AC}^{-n} \mathcal{H}_{F;k}^{(n)}$. Using the Floquet-Magnus expansion, or Baker-Campbell-

Hausdorff formula [74], we can combine the unitaries in Eq. (D3) into one unitary $\exp(-i \sum_j t_{\text{acq}} \tilde{\mathcal{H}}_{\text{acq}, k+j} + 2\pi/N I_x)$, up to an error of order $\mathcal{O}(t_{\text{acq}} J, 2\pi/N)$, where J is the energy scale of the Hamiltonian.

Since the kick angle $2\pi/N$ is large ($\sim \mathcal{O}(1)$), also terms higher order in $2\pi/N$ (i.e., $\mathcal{O}(t_{\text{acq}} J, j), (2\pi/N)^n$), contribute to the leading-order expression for $\tilde{\mathcal{H}}_{F,k}^{(0)}$ in T_{AC} . Instead, we can account for contributions of the SL pulse to the Floquet Hamiltonian exactly, by writing Eq. (D3) in the toggling frame as

$$\tilde{U}_{F,k} = \prod_{j=1}^N \tilde{U}_{\text{acq}, k+j} \tilde{U}_{\text{SL}} = \tilde{U}_{\text{SL}} \prod_{j=1}^N \tilde{U}_{\text{SL}}^{-j} \tilde{U}_{\text{acq}, k+j} \tilde{U}_{\text{SL}}^j, \quad (\text{D6})$$

which is obtained by introducing N identities, $\mathbf{1} = U_{\text{SL}}^{-j} U_{\text{SL}}^j$.

This leads to an average Hamiltonian, which is valid up to $\mathcal{O}(T_{\text{AC}})$ -corrections, and reads as

$$\begin{aligned} \frac{t_{\text{acq}}}{\tau} \tilde{\mathcal{H}}_{F,k}^{(0)} &= \sum_{j=1}^N \tilde{U}_{\text{SL}}^{-j} \tilde{\mathcal{H}}_{\text{acq}, k+j} \tilde{U}_{\text{SL}}^j \\ &= \tilde{\mathbf{w}}_k(\alpha, \phi_{\text{AC}}) \cdot \mathbf{I} \\ &\quad + (1 - 3 \sin^2 \alpha) \sum_{n < m} b_{nm} \left(\frac{3}{2} \tilde{H}_{\text{ff}} - \mathbf{I}_n \cdot \mathbf{I}_m \right), \end{aligned} \quad (\text{D7})$$

with the flip-flop term $\tilde{\mathcal{H}}_{\text{ff}} = I_{jz} I_{kz} + I_{jy} I_{ky}$, and where

$$\tilde{\mathbf{w}}_k(\alpha, \phi_{\text{AC}}) = \left(\sin(\alpha) \delta + \frac{\delta \vartheta}{\tau} \right) \hat{\mathbf{x}} + \tilde{\mathbf{u}}_k(\alpha, \phi_{\text{AC}}), \quad (\text{D8})$$

with

$$\begin{aligned} \tilde{\mathbf{u}}_k(\alpha, \phi_{\text{AC}}) &= B_{\text{AC}} \cos \alpha \sum_{n=1}^N (-\sin n\vartheta \hat{\mathbf{y}} \\ &\quad + \cos n\vartheta \hat{\mathbf{z}}) f_{n+k}(\phi_{\text{AC}}). \end{aligned} \quad (\text{D9})$$

A priori, it is unclear whether the sum in Eq. (D9) would yield a nonzero number, since both terms under the sum vanish individually, i.e., $\sum_n f_n = 0$ and $\sum_{n=1}^N (-\sin n\vartheta \hat{\mathbf{y}} + \cos n\vartheta \hat{\mathbf{z}}) = 0$. However, for the special case $\vartheta \approx 2\pi/N$ they interfere constructively to yield a finite contribution.

To describe this more clearly, let us consider the exemplary case of $N = 4$, sketched in Fig. 19, for $\vartheta \approx \pi/2$ and $\vartheta \approx \pi$. Both cases are sketched in Fig. 19(c)(i), with the colored and black vectors, respectively. First, we note that by symmetry, and regardless of ϑ , $f_1 = -f_3$ and $f_2 = -f_4$, due to the periodicity of the sine function [see Fig. 19(a)]. Let us first consider the case of $\vartheta \approx \pi/2$. Defining the vectors $\mathbf{v}_n(\vartheta) = (-\sin n\vartheta \hat{\mathbf{y}} + \cos n\vartheta \hat{\mathbf{z}})$, we then have the quantities

$$\mathbf{a}_{1,3} = \mathbf{v}_{1,3}(\pi/2) = \pm \hat{\mathbf{y}}, \quad \mathbf{a}_{2,4} = \mathbf{v}_{2,4}(\pi/2) = \mp \hat{\mathbf{z}},$$

and following Eq. (D9), the emergent field is then given by $\tilde{\mathbf{u}}_k(\alpha, \phi_{\text{AC}}) = B_{\text{AC}} \cos \alpha \sum_n f_{n+k}(\phi_{\text{AC}}) \mathbf{a}_n$. Now, the two pairs of weights cancel each other out, i.e., $f_1 = -f_3$ and $f_2 = -f_4$, and are associated with antiparallel vectors; see Fig. 19(b). This leads to an enhancement instead of a cancellation, and hence $\tilde{\mathbf{u}}_{k=0}(\vartheta = \pi/2) = 2f_1 \hat{\mathbf{y}} - 2f_2 \hat{\mathbf{z}}$ [see Fig. 19(c)(i)]. The

other vectors $\tilde{\mathbf{u}}_k$ ($k = 1, 2, 3$) are simply related to the first one by micromotion-induced rotation about the $\hat{\mathbf{x}}$ axis.

However, for the case of $\vartheta \approx \pi$, we have

$$\mathbf{a}_{1,3} = \mathbf{v}_{1,3}(\pi) = \hat{\mathbf{z}} \quad \mathbf{a}_{2,4} = \mathbf{v}_{2,4}(\pi) = -\hat{\mathbf{z}},$$

In this case, the interference is destructive yielding $\tilde{\mathbf{u}}_{k=0}(\vartheta = \pi) = (f_1 + f_3 - f_2 - f_4) \hat{\mathbf{z}} = \mathbf{0}$ [see black vector in Fig. 19(c)(i)].

From Eq. (D7) it follows directly that the N different stroboscopic Floquet Hamiltonians are related by a rotation about the $\hat{\mathbf{x}}$ axis: $\tilde{\mathcal{H}}_{F,k'}^{(0)} = \tilde{U}_{\text{SL}}^{k'-k} \tilde{\mathcal{H}}_{F,k}^{(0)} \tilde{U}_{\text{SL}}^{k-k'}$. Notice that the leading-order Floquet Hamiltonians $\tilde{\mathcal{H}}_{F,k}^{(0)}$ only differ in their single-particle terms $\tilde{\mathbf{w}}_k(\alpha, \phi_{\text{AC}}) \cdot \mathbf{I}$. Moreover, in the absence of an AC drive, the Hamiltonians $\tilde{\mathcal{H}}_{F,k}^{(0)}(B_{\text{AC}} = 0)$ become k -independent and are invariant under global rotations about the x axis, i.e., $[\tilde{\mathcal{H}}_{F,k}^{(0)}(B_{\text{AC}} = 0), I_x] = 0$. To sum up, introducing a finite AC drive leads to nontrivial emergent fields, but it does not affect the dipole-dipole interaction term, to lowest order in T_{AC} . We emphasize that these arguments still hold true after undoing the basis transformation V ; however, the conserved axis (for $B_{\text{AC}} = 0$) is rotated into $\hat{\mathbf{n}} = \cos(\alpha) \hat{\mathbf{x}} + \sin(\alpha) \hat{\mathbf{z}}$.

These expressions are derived in the simplified tilde basis that does not account for the tilt of the trajectories away from the $\hat{\mathbf{x}}$ axis on account of SL pulse detuning. In order now to write down the average Hamiltonian in the original rotating frame, we carry out the transformation $\mathcal{H}_{F,k}^{(0)} = V^\dagger \tilde{\mathcal{H}}_{F,k}^{(0)} V$, resulting in

$$\begin{aligned} \frac{t_{\text{acq}}}{\tau} \mathcal{H}_{F,k}^{(0)} &= \mathbf{w}_k(\alpha, \phi_{\text{AC}}) \cdot \mathbf{I} \\ &\quad + (1 - 3 \sin^2 \alpha) \sum_{n < m} b_{nm} \left(\frac{3}{2} H_{\text{ff}} - \mathbf{I}_n \cdot \mathbf{I}_m \right), \end{aligned} \quad (\text{D10})$$

with the transformed flip-flop Hamiltonian

$$\begin{aligned} H_{\text{ff}} &= \cos^2(\alpha) I_{jz} I_{kz} + \sin^2(\alpha) I_{jx} I_{kx} + I_{jy} I_{ky} \\ &\quad - \cos(\alpha) \sin(\alpha) (I_{jx} I_{kz} + I_{jz} I_{kx}), \end{aligned} \quad (\text{D11})$$

and the single-particle contributions

$$\mathbf{w}_k(\alpha, \phi_{\text{AC}}) = \mathbf{v}(\alpha) + \mathbf{u}_k(\alpha, \phi_{\text{AC}}), \quad (\text{D12})$$

where

$$\mathbf{v}(\alpha) = \left(\sin(\alpha) \delta + \frac{\delta \vartheta}{\tau} \right) (\cos(\alpha) \hat{\mathbf{x}} + \sin(\alpha) \hat{\mathbf{z}}),$$

$$\begin{aligned} \mathbf{u}_k(\alpha, \phi_{\text{AC}}) &= B_{\text{AC}} \cos \alpha \sum_{n=1}^N [-\sin n\vartheta \hat{\mathbf{y}} \\ &\quad + \cos n\vartheta (\cos(\alpha) \hat{\mathbf{z}} - \sin(\alpha) \hat{\mathbf{x}})] f_{n+k}(\phi_{\text{AC}}) \end{aligned} \quad (\text{D13})$$

to leading order in the AC period T_{AC} .

Since undoing the basis transformation V hides the simplicity of the result and undoing the transformation is straightforward, we will hereinafter carry out most of the analysis in the transformed frame and only undo the transformation for the final expressions.

We emphasize that, due to the finite detuning ($\delta \neq 0$), the vectors $\mathbf{w}_k$ are not centered around the $\hat{\mathbf{x}}$ axis. Instead, the points measured on the Bloch sphere are centered around $\hat{\mathbf{n}} =$

$\cos(\alpha)\hat{\mathbf{x}} + \sin(\alpha)\hat{\mathbf{z}}$, and thus feature a finite $\hat{\mathbf{z}}$ tilt that moves the midpoint of the polygon into the lower or upper half of the Bloch-sphere as sketched in Fig. 19(c). This is important for obtaining closed shapes as we are only able to measure the modulus of the $\hat{\mathbf{z}}$ -polarization in the experiment (see Fig. 19).

3. Properties of the state in the prethermal plateau

The leading-order Floquet Hamiltonian $\tilde{\mathcal{H}}_{F;k}^{(0)}$ describes the steady state of the system in the prethermal plateau [75]. When measured at times $nT_{AC} + k\tau$ for some integers $n, k < N$, we thus anticipate the system to prethermalize to a state that is well-described by a prethermal density matrix,

$$\tilde{\rho}_{F;k} = \exp(-\beta\tilde{\mathcal{H}}_{F;k}^{(0)})/Z, \quad (\text{D14})$$

with $Z = \text{Tr} \exp(-\beta\tilde{\mathcal{H}}_{F;k}^{(0)})$. Due to the unitarity of the time evolution, the temperature β_k^{-1} is determined by the initial energy density $\varepsilon_k[\tilde{\rho}_0] = \text{Tr}\{\tilde{\rho}_0\tilde{\mathcal{H}}_{F;k}^{(0)}\}/L \stackrel{!}{=} \text{Tr}\{\tilde{\rho}_{F;k}\tilde{\mathcal{H}}_{F;k}^{(0)}\}/L$ which matches the energy density of the state evolved in the prethermal plateau. For an initial state with polarization μ along the x -direction $\tilde{\rho}_0 \propto (\mathbf{1} + \mu I_x)$, using $\sin(\alpha) = \delta/\sqrt{\delta^2 + \Omega^2}$, we find that the initial energy density

$$\varepsilon_k = \text{Tr}\{\tilde{\rho}_0\tilde{\mathcal{H}}_{F;k}^{(0)}\}/L = \mu|(\delta^2/\sqrt{\delta^2 + \Omega^2}) + (\delta\vartheta/\tau)|, \quad (\text{D15})$$

in general, does not vanish, as long as either $\delta\vartheta$ or δ are finite. Equation (D15) is a direct result of the tracelessness of the spin operators $\text{Tr}(I_a) = \text{Tr}(I_{na}I_{mb}) = 0$, for $a, b = x, y, z$ and $n \neq m$, and $\text{Tr}(I_a I_a)/Z = L$.

Notice that the initial polarization, and with it the inverse temperature, is relatively small; this justifies the high-temperature approximation $\tilde{\rho}_{F;k} \approx (\mathbf{1} - \beta\tilde{\mathcal{H}}_{F;k}^{(0)})/Z$. Within the high-temperature approximation, using the relation $|\mathbf{w}_k|^2 = |\tilde{\mathbf{w}}_k|^2$, for the inverse temperature we find

$$\beta_k = -\frac{\varepsilon_k}{h_{\text{dd}}^2 + |\mathbf{w}_k|^2}. \quad (\text{D16})$$

Note that the (inverse) temperature at which the system prethermalizes is set by the relative phase ϕ_{AC} between the AC and the SL pulse drives; e.g., for $\phi_{AC} = 0$, we have $\beta = \beta_{k=0}$, etc. We stress that, once the system has reached the prethermal plateau, its state has a single well-defined temperature throughout the entire micromotion dynamics.

In Eq. (D16),

$$h_{\text{dd}}^2 = 2(1 - 3\sin^2\alpha)^2 \sum_{n < m} b_{nm}^2/L \propto J^2 \quad (\text{D17})$$

is an energy scale associated with the dipole-dipole coupling. In general, h_{dd} is finite also in the thermodynamic limit since $b_{nm}^2 \propto 1/|r_{nm}|^6$ is sufficiently short-ranged in three dimensional space for the sum above to converge. Here, r_{nm} is the internuclear position vector. However, the precise value of h_{dd} depends on microscopic details and cannot be accessed easily.

To summarize, as stated in the main text, it follows that the polarization in the prethermal plateaus is determined by

$$\mathbf{M}_k = \text{Tr}\{\mathbf{I}\rho_{F;k}\}/L = \frac{-\varepsilon_k}{h_{\text{dd}}^2 + |\mathbf{w}_k|^2} \mathbf{w}_k, \quad (\text{D18})$$

which scales linearly with the AC amplitude, $\mathbf{M}_k \propto B_{AC}$, for small amplitudes compared to the dipole-dipole coupling, $B_{AC}/h_{\text{dd}} \ll 1$.

To characterize the measured prethermal polygon shapes, let us introduce two metrics: the linear size ℓ and the excursion angle Φ away from the spin-lock axis. To quantify the size of the shapes independent of the number of measured points N , we note that all points lie on a circle in 3D, and hence we can use the radius of this circle to define the linear size ℓ of the polygons:

$$\ell = |\mathbf{w}_{k;\parallel}| = |\mathbf{w}_k - \hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{w}_k)|. \quad (\text{D19})$$

As the corners of the polygon, $\mathbf{w}_k$, all lie in one plane, we can extract the in-plane components $\mathbf{w}_{k;\parallel} = \mathbf{w}_k - \mathbf{w}_{k;\perp}$ by subtracting the center point $\mathbf{w}_{k;\perp} = \hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{w}_k)$ of the polygon, where $\hat{\mathbf{n}} = \cos(\alpha)\hat{\mathbf{x}} + \sin(\alpha)\hat{\mathbf{z}}$ is the unit-vector perpendicular to the plane.

In addition, let us define the excursion angle

$$\Phi = 2 \arctan\left(\frac{\ell}{|\mathbf{w}_{k;\perp}|}\right). \quad (\text{D20})$$

The excursion angle is a measure for the size of the deviation of the polygon points from the axis $\hat{\mathbf{x}}$ which is conserved in the absence of the AC drive. We note that, for regular shapes, these two measures are single valued, i.e., Φ and ℓ should be independent of the index k .

Using Eq. (D18) it is straightforward to show that

$$\ell \propto \frac{\varepsilon_k B_{AC}}{h_{\text{dd}}^2 + |\mathbf{w}_k|^2} \quad (\text{D21})$$

and

$$\tan\left(\frac{\Phi}{2}\right) = \varepsilon_k \frac{|\mathbf{u}_k|}{|\mathbf{v}(\alpha)|} \propto B_{AC}. \quad (\text{D22})$$

4. Theory of the quench dynamics

The transient dynamics when turning on and off the AC field are highly model dependent. Nevertheless, we can make qualitative predictions about the behavior of the system by considering the thermal properties of the density matrices before and after the quench. We will, in particular, consider three scenarios, (i) initial opening of the shapes, (ii) closing of the shapes, and (iii) the combination of closing and reopening, corresponding to the scenarios depicted in Figs. 4 and 10. We will restrict the following analysis to “square” shapes, for simplicity.

In the following analysis we will analyze the states ρ_f the system is expected to thermalize to after each quench, given the properties of the state ρ_i before each quench. Following the discussion in the main text, we will further denote states for which the AC field is on (off) by a on (off) subscript.

The initial opening $\tilde{\rho}_{\text{init}} \rightarrow \tilde{\rho}_{\text{on},f}$ has already been described in detail in Appendix D3. For the closing of the shapes, $\tilde{\rho}_{\text{on},i} \rightarrow \tilde{\rho}_{\text{off},f}$, we point out the special role of the $\hat{\mathbf{x}}$ -polarization conservation, such that the thermal state is characterized by both a temperature and a polarization. To account for the conservation law we introduce an additional Lagrange multiplier γ in the prethermal ensemble

$$\tilde{\rho}_{\text{off},f} = \exp(-\beta_{\text{off},f}[\tilde{\mathcal{H}}_F^{(0)}(B_{AC}=0) - \gamma I^x])/Z. \quad (\text{D23})$$

The inverse temperature $\beta_{\text{off},f}$ and the new parameter γ are determined by the initial energy $\text{Tr}\{\tilde{\rho}_{\text{on},i}\tilde{\mathcal{H}}_F^{(0)}\} = \text{Tr}\{\tilde{\rho}_{\text{off},f}\tilde{\mathcal{H}}_F^{(0)}\}$ and polarization $\text{Tr}\{I_x\tilde{\rho}_0\} = \text{Tr}\{I_x\tilde{\rho}_F(B_{AC}=0)\}$, respectively.

The initial state $\tilde{\rho}_{\text{on},i}$ is given by the thermal state before the quench, i.e., $\tilde{\rho}_{\text{on},i} = \tilde{\rho}_{F;0}(\phi_c) = \exp(-\beta\tilde{\mathcal{H}}_{F;0}^{(0)}(\phi_c))/Z$, introduced in Appendix D 3, where the phase at closing is determined by the time t_c at which the closing happens via $\phi_c = \phi_{AB} + (\omega_{AC}t_c) \bmod(2\pi)$.

Combining the high temperature approximation $\tilde{\rho}_{\text{on},i} \approx (\mathbf{1} - \beta_{\text{on},i}\tilde{\mathcal{H}}_{F;0}^{(0)}(\phi_c))/Z$ and $\tilde{\rho}_{\text{off},f} \approx (\mathbf{1} - \beta_{\text{off},f}[\tilde{\mathcal{H}}_F^{(0)}(B_{AC}=0) - \gamma I_x])/Z$, the temperature and polarization after the quench are immediately given by

$$\beta_{\text{off},f} = \beta_{\text{on},i} \quad \text{and} \quad \gamma = 0. \quad (\text{D24})$$

We emphasize that this does not imply the equality of the two states, since $\tilde{\mathcal{H}}_F^{(0)}(B_{AC}=0) \neq \tilde{\mathcal{H}}_F^{(0)}(B_{AC} \neq 0)$. However, our result suggests that the closing quench merely leads to a decay of all components orthogonal to the preserved axis $\hat{x}$. In this respect, note also that the final state, after closing the shape, has a finite dipolar order. In contrast, the initial state $\tilde{\rho}_{\text{init}} \propto I_x$ has a vanishing dipolar order.

For these reasons, reopening the shapes, $\rho_{\text{on},i} \rightarrow \rho_{\text{on},f}$ is different from the initial opening of the shapes. In particular, the temperature after reopening is given by

$$\beta_{\text{on},f} = \beta_{\text{on},i} \frac{h_{\text{dd}}^2 + \mathbf{v}(\alpha) \cdot \mathbf{w}_k}{h_{\text{dd}}^2 + |\mathbf{w}_k|^2}, \quad (\text{D25})$$

where $\beta_{\text{on},i}$ is the temperature before the quench. The fraction on the right-hand side of Eq. (D25) is always smaller than 1, as $|\mathbf{w}_k| \geq |\mathbf{v}(\alpha)|$, with equality only at $B_{AC} = 0$.

Therefore, combining Eqs. (D18), (D24), and (D25), we find that the total polarization $|\mathbf{M}_{\text{on},f}|$, after closing and reopening, is given by

$$|\mathbf{M}_{\text{on},f}|/|\mathbf{M}_{\text{on},i}| = \beta_{\text{on},f}/\beta_{\text{on},i} = 1 - \sigma. \quad (\text{D26})$$

Here $|\mathbf{M}_{\text{on},i}|$ is the total polarization prior to closing, and $\sigma = (h_{\text{dd}}^2 + \mathbf{v}(\alpha) \cdot \mathbf{w}_k)/(h_{\text{dd}}^2 + |\mathbf{w}_k|^2)$. Hence, with each closing and reopening, the amplitude of the polarization is reduced by a factor $(1 - \sigma)$. However, each closing and reopening process results only in a tiny loss, since $\sigma \ll 1$ in the experimentally relevant regime $B_{AC} \ll h_{\text{dd}}$.

APPENDIX E: NUMERICAL SIMULATIONS

Our experimental and analytical results are complemented by exact numerical simulations performed on a system of L interacting ^{13}C nuclear quantum spins positioned on a pseudorandom graph.

1. Numerical model

For the design of pseudorandom graphs we respect the following condition [14]: each spin of the graph is required to have at least one nearest neighbor within a distance r_{max} while no other spin is closer than r_{min} . This ensures a certain average spin density and, importantly, avoids weakly coupled outliers. While such weakly coupled spins are in principle expected to emerge to some extent in the experimental system, their influence on the many-body dynamics is negligible. In particular,

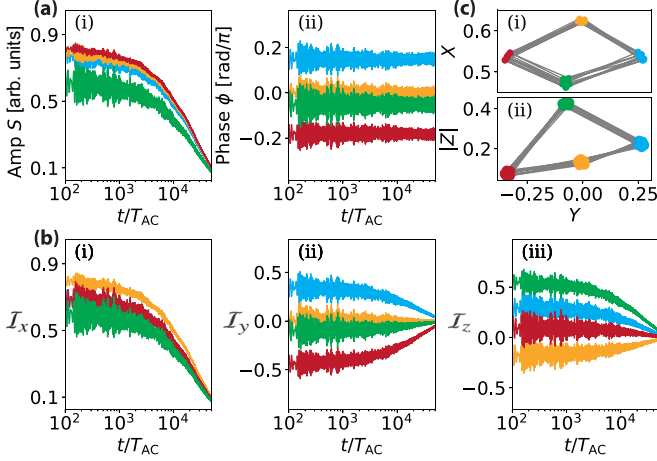
from a numerical perspective where only a relatively small number of interacting spins can be simulated, outliers are wastefully problematic as each of them reduces the effective interacting Hilbert space by a factor of 2.

The pseudorandom graphs are generated iteratively: first, we randomly propose a new spin position j and check if (i) the interspin vectors with all other spins k on the graph satisfy $|\vec{r}_{jk}| > r_{\text{min}}$, while (ii) at least one interspin vector satisfies $|\vec{r}_{jk}| < r_{\text{max}}$. If (i) and (ii) are satisfied, then we accept the proposal and update the random graph with the spins at position j . Otherwise, we discard the proposed spin. We repeat this procedure until a graph of L spins is constructed. The simulations are performed with $L = 14$, $r_{\text{min}} = 0.9$ and $r_{\text{max}} = 1.1$; here r_{min} and r_{max} are given in units of $\sqrt[3]{\mu_0\hbar\gamma_n^2}$.

For numerical simulations, unless stated explicitly otherwise, we initialize the system in a $\hat{x}$ -polarized pure product state $|\psi_0\rangle = \bigotimes_{j=1}^L \frac{1}{\sqrt{2}}(|\uparrow_j\rangle + |\downarrow_j\rangle)$ and perform numerically exact time evolution using the protocol of Eq. (3). Due to the critical long-range interaction strength, the characteristic energy scales of the model are not immediately obvious (the energy density integral diverges in three spacial dimensions, which indicates the necessity to impose a lattice cutoff scale). However, they can be estimated from the dynamics of a free-induction decay: to this end, the initial state $|\psi_0\rangle$ is evolved under $\mathcal{H}_{\text{dd}}$ which induces a decay of the initial $\hat{x}$ -polarization. Eventually, this provides an energy scale $J = 1/\tau_d$ where τ_d is the timescale on which the $\hat{x}$ -polarization decays to $1/e$ of its initial value as the system approaches equilibrium. We note that the value of J obtained in this way can only serve as a rough estimate of energy scales, as different initial states might lead to different values of J .

To minimize detrimental finite-size effects and increase the ergodic properties of the drive we add a small uniformly distributed random noise $\delta t_{\text{acq}} \in [-0.05t_{\text{acq}}, 0.05t_{\text{acq}}]$ to the time duration t_{acq} , i.e., $t_{\text{acq}} \rightarrow t_{\text{acq}} + \delta t_{\text{acq}}$ [76,77] which results in a slightly different unitary for each driving cycle. Such a “noisy” driving protocol can remove left-over synchronization effects emerging from the discreteness of the many-body spectrum. However, since the noise term δt_{acq} breaks the periodicity of the drive, one needs to carefully examine that no additional physics, such as an extra contribution to heating, is induced due to finite δt_{acq} . For the system under investigation, this has been demonstrated in Ref. [14].

Finally, let us present some additions to the data presented in the main text, i.e., in Figs. 2 and 3. In Fig. 20, we present the full numerical data for the simulated time evolution partially depicted in Fig. 2(b). In particular, the simulated data also leads to the square shapes when projected to the $\hat{x}$ - $\hat{y}$ or $\hat{y}$ - $\hat{z}$ plane. In addition to the excursion angle Φ and linear scale ℓ shown in Figs. 3(c) and 3(d), we show the corresponding raw data and decay times as functions of the AC amplitude in Fig. 21. Let us in particular point out the increase in the lifetime with increasing AC Amplitude by a factor of up to 2, suggested by the numerical simulation. This is not observed in the experiment and obtaining analytical expressions for the decay time are beyond reach for our nonintegrable long-range interacting systems. Notice however that the lifetime is only affected by a factor of 2 over a large range of AC amplitudes and that the impact of finite-size effects on the various quantities is not well-understood.

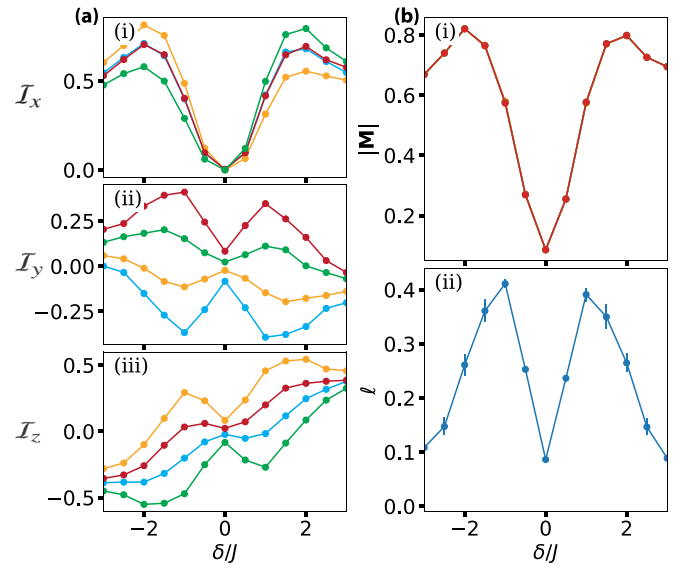
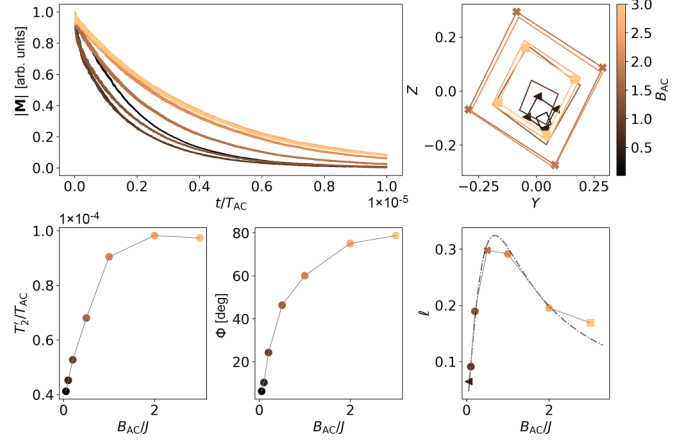


2. Effect of detuning and imperfect pulses on observed shapes

In the main text we discussed two imperfections of the SL pulse drive: the detuning δ from the Larmor frequency ω_L and the deviation in the pulse angle $\delta\vartheta = \vartheta - 2\pi k/N$. Here, we show that they can lead to an enhancement of the signal. In fact, they are essential for reproducing the experimental observations. Eventually, we also investigate the related question of having noise in the kick sequence.

We start with the detuning δ . Let us stress again that, due to the large scale separation $\omega_L/J \leq 10^{-5}$, even a small deviation (0.01%) can lead to a comparably large detuning $\delta \leq J$. Note that the detuning affects the direction of the kicks $\hat{x} \rightarrow \hat{n} = \cos(\alpha)\hat{x} + \sin(\alpha)\hat{z}$, cf. Eq. (D4). Therefore, the obtained shapes lie on circles which are centered around the $\hat{n}$ axis. Note that $\hat{n}$ has a finite $\hat{z}$ contribution, $\hat{n}_z \propto \delta$, which leads to a finite $\hat{z}$ tilt, as can also be observed in the simulations in Fig. 22. Therefore, the detuning is crucial for explaining the experimentally observed $\hat{z}$ tilt.

Moreover, the detuning, together with the imperfection in the pulse strength $\delta\vartheta$, leads to a finite $\hat{x}$ -polarization, $M_x \propto \sin(\alpha) + \delta\vartheta/\tau$, in the leading-order Floquet Hamiltonian $\mathcal{H}_{F;k}^{(0)}$, cf. Eq. (5). This leads to an increase in the $\hat{x}$ -polarization strength, as can also be observed in Figs. 22(a)(i) and 23(a)(i). In addition, as mentioned in Appendix D 3, it is vital that δ or $\delta\vartheta$ are finite for the initial state to have a finite energy density ϵ , and thus also a finite temperature $|\beta| \propto |\sin(\alpha) + \delta\vartheta/\tau| > 0$. Such an increase in temperature



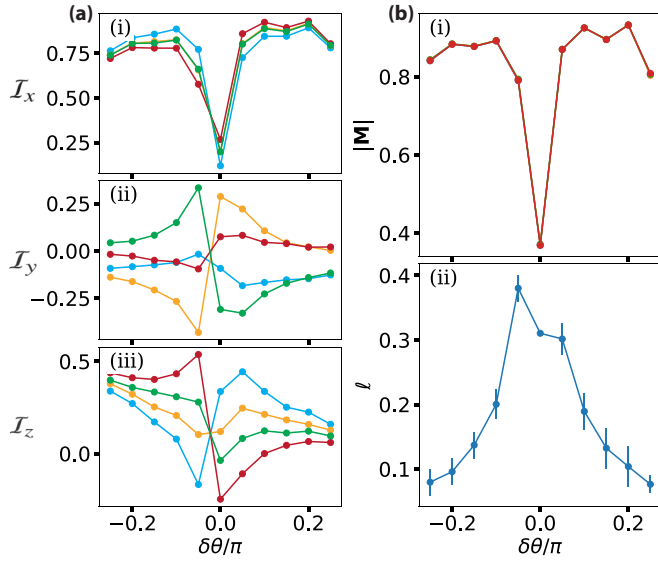


FIG. 23. Simulations of effect of imperfect pulse duration. (a) polarization components of M in (i) $\hat{x}$, (ii) $\hat{y}$, and (iii) $\hat{z}$ direction. (b) Total polarization (i) $|M|$ and linear scale (ii) ℓ as a function of the imperfectness $\delta\vartheta$ in the angle $\vartheta = 2\pi/4 + \delta\vartheta$. Different colors indicate the $N = 4$ different plateaus. Data is extracted after 1000 pulses and averaged over 5 disorder realizations for a system of $L = 14$ spins with fixed AC amplitude $B_{AC}/J = 0.5$ and detuning $\delta/J = 1$, and variable accumulated pulse angle $\vartheta = \pi/2 + \delta\vartheta$. Further parameters are as in Fig. 20.

is correlated with the increase in $\hat{y}$ - and $\hat{z}$ -polarizations in Figs. 22 and 23, cf. Eq. (D18).

Note that the robustness of the shapes with respect to imperfection in the kicks, i.e., for $\delta\vartheta \neq 0$, also demonstrates

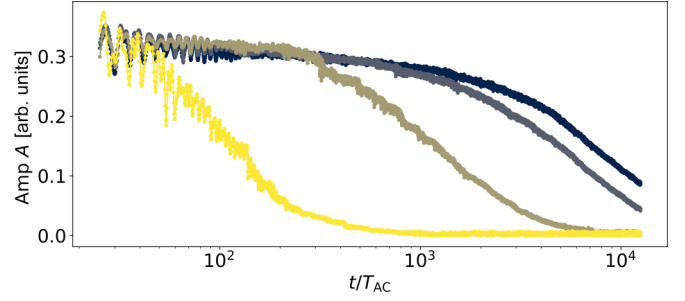


FIG. 24. Simulation: Noisy Pulses. Total polarization $|M| = \sqrt{M_x^2 + M_y^2 + M_z^2}$ as a function of time for disordered kick sequences with varying strength η . Decrease in the lifetime of the prethermal plateaus with increasing disorder is observed. Data is obtained for $L = 14$ spins at fixed detuning $\delta/J = 1.0$, AC amplitude $B_{AC}/J = 0.5$ and fluctuating accumulated phase $\vartheta = \pi/2(1 + \zeta)$ with varying uniform fluctuations $\zeta \in [-\eta, \eta]$, $\eta = 0, \dots, 3.16\%$ (dark to light). Further parameters are as in Fig. 20.

that the shapes are not simply a single-particle effect but are stabilized by the many-body interactions.

Finally, let us emphasize that we numerically do not observe a significant change in the lifetime when varying the detuning δ or the angle $\delta\vartheta$.

To explore this behavior, we simulate noisy pulses $\vartheta = (1 + \zeta) \times 2\pi/4$ for the special case of $N = 4$, where ζ are random numbers that differ for every pulse uniformly distributed in the interval $[-\eta, \eta]$ (with η being the noise strength). We observe a strong decrease in the lifetime of the prethermal plateau with increasing η ; see Fig. 24. This result is not surprising as the periodic nature of the drive is lost with increasing randomness.

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