

Observation of Hierarchy of Hilbert Space Ergodicities in the Quantum Dynamics of a Single Spin

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Ergodicity, the property that all allowed configurations are explored over time, plays a pivotal role in explaining the equilibrium behavior of classical dynamical systems. Yet, such a property is typically precluded in quantum systems owing to stationary energy eigenstates. However, recent theoretical works have argued that ergodic explorations of the Hilbert space, occurring at varying levels as measured by statistical pseudorandomness of the time-evolved states, may happen for aperiodic driven quantum systems. Here, we experimentally investigate the hierarchy of Hilbert-space ergodicities (HSEs) achievable in the dynamics of a single spin. Through subjecting a nitrogen-vacancy center in diamond to various time-dependent modulations and continuously monitoring the spin trajectories with full state tomography, different degrees of HSE were observed, ranging from no HSE in a time-periodic drive, to partial HSE in a smoothly kicked time-quasiperiodic drive, to complete HSE in an aperiodic Fibonacci drive. We formulate a theoretical understanding of the increasing levels of HSE by attributing them to increasing levels of complexity associated with the drive sequences. Our Letter provides the first unambiguous experimental evidence of Hilbert space ergodicity, promoting deeper investigations into the mechanisms and fine-grained levels with which closed quantum systems reach equilibrium.

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Ergodicity in classical systems refers to the dynamical property that a system explores its allowed phase space uniformly over time (with respect to a natural invariant measure), regardless of initial state [1–3]. It forms the cornerstone of statistical physics, explaining how equilibrium arises from complex dynamics [4,5]. In the quantum realm, on the other hand, the idea of ergodicity is formulated rather differently [6–11]. Owing to the fact that energy eigenstates of quantum Hamiltonians are stationary, an initial-state-independent dynamical notion of ergodicity is not possible, and instead, quantum ergodicity is typically formulated in terms of the energy eigenstates’ statistical similarity to random vectors [12–16]. These include Berry’s conjecture in single-particle chaotic

systems [17,18], and the eigenstate thermalization hypothesis in many-body interacting systems [19–27].

Recent theoretical works, however, have argued that a dynamical formulation of ergodicity may be recovered in quantum systems driven by Hamiltonians with *aperiodic time dependencies*, where energy eigenstates may not exist [28–31]. As a simple example, a quantum spin driven by randomly fluctuating fields can be shown to completely and uniformly explore its Hilbert space over time, i.e., achieve “complete Hilbert space ergodicity” (CHSE). However, Ref. [28] showed more nontrivially that a quantum system driven by a nonrandom but aperiodic sequence of kicks called the Fibonacci drive also achieves CHSE. Subsequent works [29,30] have provided conditions when CHSE, and more generally, when finite levels of Hilbert-space ergodicity (HSE) may be achieved, depending on the drive. While a recent work [32] found signatures of HSE in the dynamics of a Rydberg atom quantum simulator (in the statistics of measured computational bitstrings over time), a direct experimental observation of HSE and its hierarchy—which necessitates probing the *full* temporal distribution of quantum states over the Hilbert space—has to date not been achieved.

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In this Letter, we provide the first unambiguous experimental observation of HSE, and explore the hierarchy of ergodicities achievable in the quantum dynamics of a single spin. Concretely, we subject a nitrogen-vacancy (NV) center in diamond to different time-dependent modulations, including a time-periodic (Floquet) drive, a smoothly kicked time-quasiperiodic drive, and the Fibonacci drive. To achieve dynamics over sufficiently long periods necessary to probe HSE, we synthesized an isotope-purified diamond sample whose spin dephasing time achieves 68 μs and developed quantum logic gates with theoretical fidelity over 99.99% to further resist noise. Through monitoring of spin trajectories with full quantum state tomography for nearly 1000 steps, we found evidence that the system does not exhibit HSE at any level for the Floquet drive; it exhibits HSE at only the first moment for the smoothly kicked quasiperiodic drive; while it exhibits behavior consistent with CHSE for the Fibonacci drive. To explain these findings, we argue that in addition to the number of tones underlying the drive (a notion of drive “complexity” elucidated by Ref. [30]) governing the degree of HSE achievable, the *regularity* of the modulations of the kick amplitudes over time is another important factor.

We proceed by introducing the theoretical framework of HSE. HSE concerns the statistical randomness of the sequence of states $\{|\psi(t)\rangle\}_{t \geq 0}$ of a d -dimensional quantum system generated in time by unitary dynamics under a drive Hamiltonian $H(t)$, called the “temporal ensemble.” It probes if the system explores its Hilbert space by comparing the statistical similarity of the temporal ensemble to the Haar ensemble of uniformly random states [33]. This can be quantified at different statistical moments $k \in \mathbb{N}$, captured by the vanishing of the trace distance $\Delta^{(k)}(T) := \frac{1}{2} \|\rho_T^{(k)} - \rho_{\text{Haar}}^{(k)}\|_1$ at late observation times T . Above, $\|\cdot\|_1$ is the trace norm [34], $\rho_T^{(k)} := 1/T \int_0^T dt (|\psi(t)\rangle\langle\psi(t)|)^{\otimes k}$ is the k th moment of the finite-time temporal ensemble constructed from initial state $|\psi(0)\rangle$, and $\rho_{\text{Haar}}^{(k)} := \int dU (|U\rangle\langle U|)^{\otimes k}$ is the k th moment of the Haar ensemble with dU the Haar measure on $\text{SU}(d)$ [36].

If $\Delta^{(k)}(T)$ vanishes when dynamics are initialized from *any* state $|\psi(0)\rangle$, then this property is termed “ k -HSE” (in practice, a small but finite threshold is allowed). k -HSE for $k \geq 2$ goes beyond the standard framework of equilibration at the level of time-averaged observables, captured by 1-HSE [28]. Indeed, the various levels form a strict hierarchy: k -HSE implies k' -HSE whenever $k > k'$ [30]. In particular, when HSE holds for all k , the system is said to exhibit *complete* Hilbert-space ergodicity (CHSE): the time-evolved states are indistinguishable from Haar random ones. It is this hierarchical nature of ergodicities that underpins one of the motivations of this Letter: for which quantum dynamics can we expect deeper levels of HSE, and how do they depend on properties of the driving Hamiltonian $H(t)$?

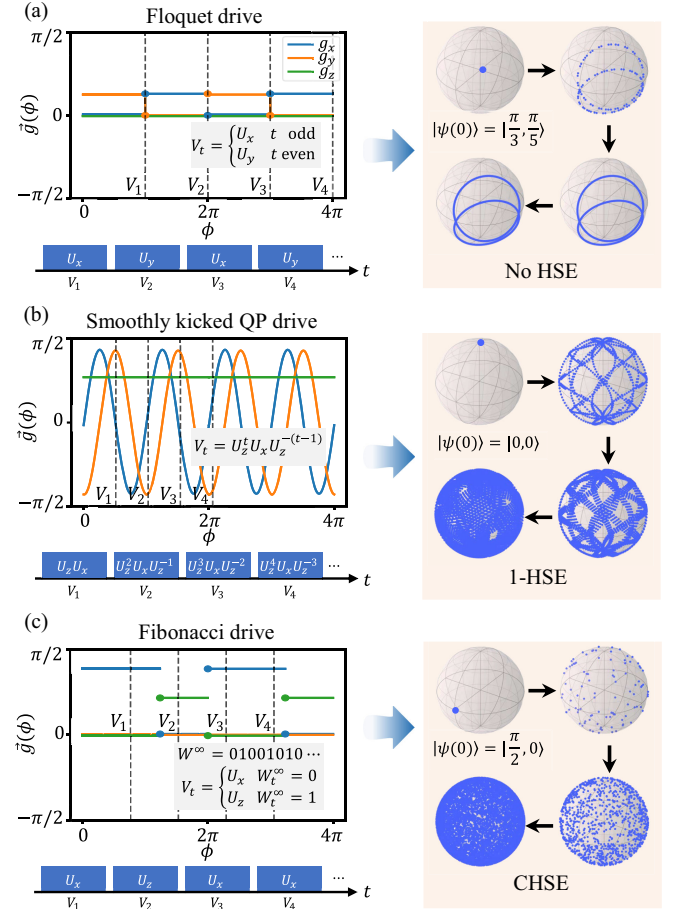


FIG. 1. Drive protocols and dynamics in the Hilbert space. (a) The Floquet, (b) smoothly kicked QP, and (c) Fibonacci drive protocols and resulting dynamics, respectively. Left panels depict the kick modulation vectors $\vec{g}(\phi)$. Evaluating them at discrete times $\omega_2 t$ (dashed vertical lines) results in kicks V_t , which build up the time-evolution operator $U(t)$. Right panels show examples of numerically simulated dynamics on the Bloch sphere starting from initial state $|\psi(0)\rangle = |\theta, \varphi\rangle$, where (θ, φ) are the polar and azimuthal angles of a qubit state.

Here, we study this question by investigating three time-dependent Hamiltonians that have distinct temporal profiles and experimentally implementing them in a qubit (so $d = 2$) realized by a single NV center. These drives are chosen to have varying forms of “complexity,” which affects the level of ergodicity achievable, discussed in more detail later. In all cases,

$$H(t) = \sum_{n \in \mathbb{N}} \delta(t - 2\pi n/\omega_1) \vec{g}(\omega_2 t) \cdot \vec{\sigma}, \quad (1)$$

where we set the first frequency component $\omega_1 = 2\pi$ for simplicity, and $\vec{\sigma}$ are standard Pauli matrices. The drives are defined by a Bloch vector $\vec{g}(\phi)$, called the *kick modulation vector*, which is a function on the circle $S^1 = [0, 2\pi] \ni \phi$, and second frequency component ω_2 . Equation (1)

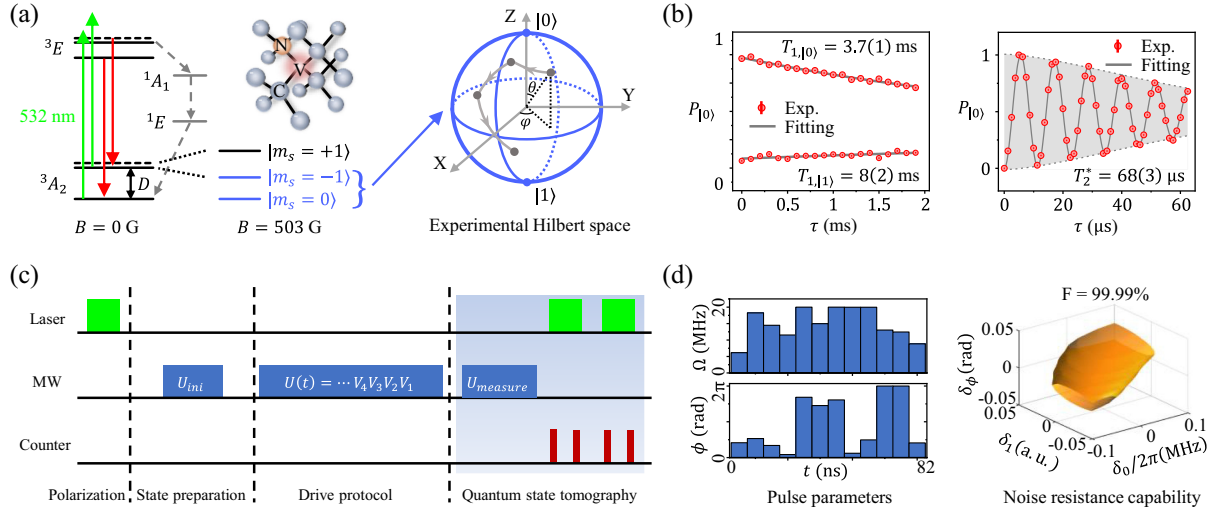


FIG. 2. NV center system and pulse scheme. (a) Atomic structure and energy levels of an NV center. An applied magnetic field splits the $|m_s = \pm 1\rangle$ states. The spin trajectories in the Bloch sphere, spanned by $|m_s = 0, -1\rangle$, were measured to explore HSE. (b) Relaxation times of the NV qubit. Error bars fall within the data points. (c) Pulse scheme to measure the spin dynamics under different drive protocols. Here, U_{ini} depends on the choice of the initial state, and different $U_{measure}$ [including I , $\mathcal{R}_X(\pi)$, $\mathcal{R}_{\pm X}(\pi/2)$, $\mathcal{R}_{\pm Y}(\pi/2)$] were implemented to perform tomography. (d) Design of high-fidelity quantum operations for example gate $U_x = e^{-i0.38\sigma_x}$. Left: amplitudes and phases of the pulses. Right: simulated noise resilience capability of the designed operation against dephasing noise δ_0 , pulse amplitude fluctuation δ_1 , and pulse phase distortion δ_ϕ .

generates a sequence of kicks $V_t = e^{-i\vec{g}(\omega_2 t) \cdot \vec{\sigma}}$ at stroboscopic times $t \in \mathbb{N}$, namely the time-evolution operator reads $U(t) = V_t \cdots V_2 V_1 = e^{-i\vec{g}(\omega_2 t) \cdot \vec{\sigma}} \cdots e^{-i\vec{g}(2\omega_2) \cdot \vec{\sigma}} e^{-i\vec{g}(\omega_2) \cdot \vec{\sigma}}$, see Fig. 1. Because of the discrete nature of dynamics, we modify the temporal ensemble definition to $\rho_T^{(k)} := 1/T \sum_{t=0}^{T-1} (|\psi(t)\rangle\langle\psi(t)|)^{\otimes k}$.

For the first drive, we set $\omega_2 = \pi$ and choose

$$\vec{g}(\phi) = \begin{cases} \theta_y \hat{y} & 0 \leq \phi < \pi, \\ \theta_x \hat{x} & \pi \leq \phi < 2\pi. \end{cases} \quad (2)$$

This yields a sequence of kicks alternating in x and y directions: $U(t) = \cdots U_y U_x U_y U_x$, where $U_\alpha = e^{-i\theta_\alpha \sigma_\alpha}$ ($\alpha = x, y, z$), i.e., a time periodic or “Floquet drive.” The second drive is defined by $\omega_2 = (1 + \sqrt{5})/2$ and

$$\vec{g}(\phi) = \frac{c}{\sin c} \begin{pmatrix} \sin 1 \cos(2\phi - \omega_2) \\ \sin 1 \sin(2\phi - \omega_2) \\ \cos 1 \sin(\omega_2) \end{pmatrix}, \quad (3)$$

with $\cos c = \cos \omega_2 \cos 1$. Note each kick V_t is equivalent to $U_z^t U_x U_z^{-(t-1)}$ with $\theta_x = 1$, and $\theta_z = \omega_2$. Since $\vec{g}(\phi)$ is a smooth function and ω_1, ω_2 are incommensurate, this is a “smoothly kicked quasiperiodic (QP) drive” of two tones. The last drive is defined by $\omega_2 = \pi(3 - \sqrt{5})$ and

$$\vec{g}(\phi) = \begin{cases} \theta_x \hat{x} & 0 \leq \phi < 2\pi - \omega_2, \\ \theta_z \hat{z} & 2\pi - \omega_2 \leq \phi < 2\pi. \end{cases} \quad (4)$$

Then, $U(t) = \cdots U_x U_z U_x U_z U_x U_x U_z U_x$, which is the Fibonacci drive [37] of Ref. [28], for which we expect CHSE.

Figure 1 shows an illustration of the three drives. As a preliminary investigation, we also show numerical plots of the trace of an example quantum state over the Hilbert space (here, the Bloch sphere) in time. One sees that the Floquet drive does not seem to explore the Hilbert space uniformly at all, while the smoothly kicked QP and Fibonacci drive do, though to apparently differing degrees. In the experimental results section, we will carefully quantify these differences as resulting in varying levels of HSE, namely no-HSE, 1-HSE, and CHSE, respectively, observations that will be further corroborated with theoretical analyses.

A single spin of an NV center in diamond was utilized in our experiments. An NV center is a point defect where a nitrogen atom replaces a carbon atom in the diamond lattice and pairs with an adjacent vacancy, as shown in Fig. 2(a) [38,39]. When negatively charged, the NV center contains six electrons and realizes a spin-1 electron spin whose energy levels are depicted in Fig. 2(a). The ground state 3A_2 is a triplet with energy levels $|m_s = 0, \pm 1\rangle$. In addition to an internal zero-field splitting of $D = 2.87$ GHz, these levels are separated by a Zeeman splitting from an externally applied magnetic field $B = 503$ G. This spectrally isolates the subspace $|m_s = 0, -1\rangle$ to be used as the qubit, which we label as $|0\rangle, |1\rangle$, respectively (the $|m_s = +1\rangle$ can be safely ignored due to the large energy mismatch and small leakage [40,41]). Initialization and readout of the spin utilize a spin-state-dependent intersystem crossing

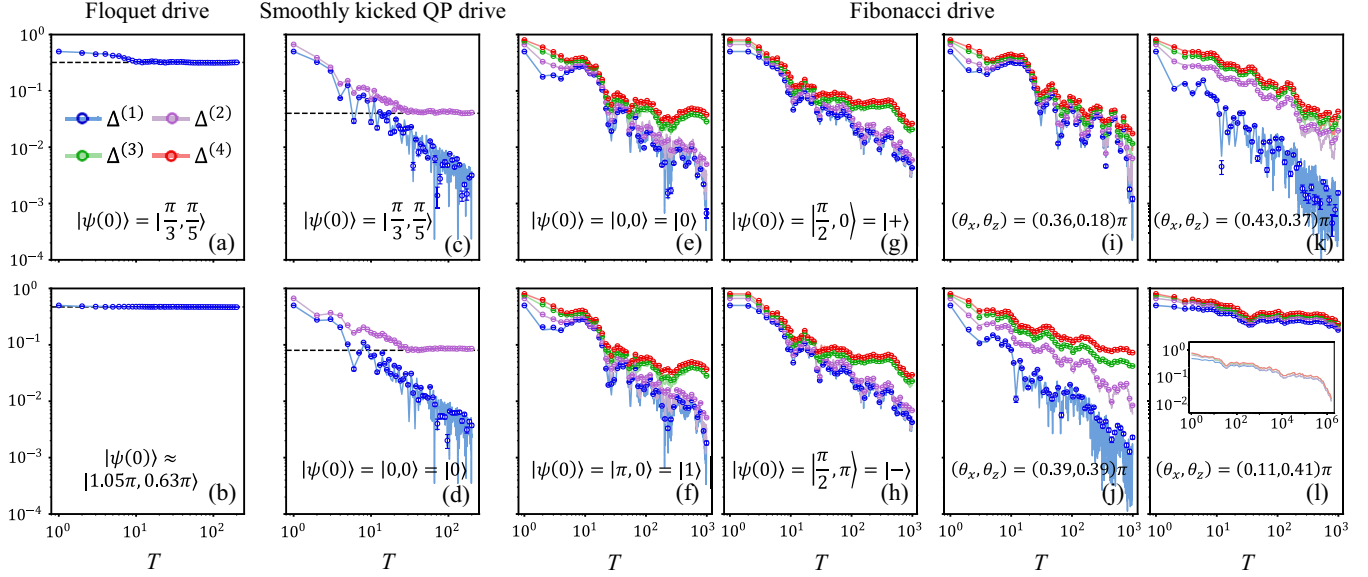


FIG. 3. Experimental results. (a),(b) Trace distance $\Delta^{(1)}$ of the Floquet drive beginning from different initial states $|\psi(0)\rangle = |\theta, \varphi\rangle$. The initial state in (b) corresponds to an eigenstate of U_F . (c),(d) Trace distances $\Delta^{(1)}$ and $\Delta^{(2)}$ plot of the smoothly kicked QP drive for various initial states. (e)–(l) Trace distances $\Delta^{(k)}$ of the Fibonacci drive for $k = 1$ to 4. In (e)–(h) we fixed the drive parameters to $(\theta_x, \theta_z) = (0.38, 0.22)\pi$ and changed the initial state, while in (i)–(l) we changed the drive parameters and fixed the initial state to $|\psi(0)\rangle = |0\rangle$. For all plots, data points with error bars are experimental results, while translucent lines of the same color are corresponding data from numerical simulations. Note that not all data points from experiments are included (to prevent information clutter); instead, they are sampled at roughly equal intervals on a logarithmic scale. Horizontal dashed lines in (a)–(d) denote plateaus.

process involving the first excited 3E and two singlet states 1A_1 and 1E , by applying a 532 nm laser pulse [42,43]. The longitudinal and transverse relaxation times of the qubit were measured and plotted in Fig. 2(b), which yield $T_{1,|0\rangle} = 3.7(1)$ ms, $T_{1,|1\rangle} = 8(2)$ ms, and $T_2^* = 68(3)$ μ s, respectively.

The pulse scheme to realize different drive protocols and measure corresponding spin dynamics is depicted in Fig. 2(c). It begins with a 532 nm laser pulse to polarize the spin to $|0\rangle$. Then, operation U_{ini} was executed to prepare different initial states $|\psi(0)\rangle$. Afterward, different target drives were applied to enact time evolution. At a desired time step, operations $U_{measure}$ followed by laser pulses were applied to perform tomography of the spin state. We note that imperfections in quantum manipulations can cause the dynamics to deleteriously deviate from idealized trajectories. To address this, we developed high-fidelity quantum logic gates for the requisite $U_{x/y}$ operations based on quantum optimal control methods [44]. We stress that in addition to dephasing and pulse amplitude noise, we also took into account pulse phase noise, which was frequently overlooked in previous studies [45,46]. In Fig. 2(d) we present an example target operation $U_x = e^{-i0.38\pi\sigma_x}$. The simulation shows the fidelity of the designed operation exceeds 99.99% within a large parameter space, numerically computed within a noise model characterizing the experimental setup. Details about the gate design and other experimental specifications are presented in Supplemental Material [47].

Our experimental findings are presented in Fig. 3. For the Floquet drive, we take $\theta_x = \theta_y = \pi/8$ and total evolution duration to be 200 time steps. Figure 3(a) shows the first-moment distance $\Delta^{(1)}$ beginning from a randomly chosen initial state (parameters indicated in the plot): there is an initial decay, but soon it plateaus to a nonzero value ≈ 0.32 . This quantifies the nonergodicity seen in the trajectory over the Bloch sphere of Fig. 1(a), and demonstrates that the Floquet drive does not exhibit 1-HSE, let alone k -HSE. The origin of the nonergodicity can be traced to stationary quasienergy eigenstates of the Floquet unitary $U_F = U_y U_x$ underlying $U(t)$. To confirm this, we repeat the same experiment beginning from one of the (numerically determined) quasienergy states of U_F ; see Fig. 3(b). As expected, $\Delta^{(1)}$ now decays even less than previously, plateauing at a higher value of ≈ 0.47 .

Results for the smoothly kicked QP drive are depicted in Figs. 3(c) and 3(d). Within the observation time interval, the first-moment distance $\Delta^{(1)}$ exhibits an overall power-law decay for all initial states. In contrast, the second-moment distance $\Delta^{(2)}$ decreases during the early stages of the evolution for both initial states, but eventually stabilizes to a nonzero value. These findings strongly suggest the emergence of 1-HSE, but not 2-HSE or higher. In the Supplemental Material [47], we confirm this by rigorously showing that the smoothly kicked QP drive forms a quantum state one-design but not two-design in dynamics, provided that ω_2 is incommensurate to both 2π and 1.

Finally, experimental results for the Fibonacci drive are illustrated in Figs. 3(e)–3(l). We first fixed the drive parameters to $(\theta_x, \theta_z) = (0.38, 0.22)\pi$ and studied dynamics starting from four different initial states: $|\psi(0)\rangle = |0\rangle, |1\rangle, |+\rangle$ and $|-\rangle$. We applied up to 987 operations and experimentally obtained up to fourth-moment trace distances [55]. Figures 3(e)–3(h) show that despite the presence of some oscillations on short timescales and transient plateaus, the overall trend is one of persistent decay for all k , consistent with a power law (understanding the origin of these oscillations and plateaus is left for future work. See, e.g., a recently uncovered mechanism of “critically slow” HSE [57]). These observations are indicative of the emergence of CHSE, in line with predictions [28]. To further ascertain universality of CHSE, we reran the experiments with different generic drive parameters $(\theta_x, \theta_z) = (0.43, 0.37)\pi, (0.36, 0.18)\pi, (0.39, 0.39)\pi$, and $(0.11, 0.41)\pi$, fixing $|\psi(0)\rangle = |0\rangle$ (avoiding fine-tuned drive parameters known to preclude CHSE [28]), see Figs. 3(i)–3(l). One sees all distances decay as T increases, with no evidence of saturation, though the decay rate varies greatly. In particular, the decay in Fig. 3(l) is exceptionally slow. Nevertheless, independent numerical simulations (inset) run for longer times show a sustained decay. Together, these observations support compellingly the CHSE property of the Fibonacci drive.

To summarize, our Letter has provided the first unambiguous experimental observation of HSE, a novel form of quantum ergodicity rooted in statistical pseudorandomness of states generated in dynamics, and explored the different settings under which varying levels of ergodicities arise. We now conclude with a discussion of the origin and implications of our findings. We ask: can the different levels of HSE seen in the drives (and more broadly in other drives) already be anticipated based on certain general physical principles?

Now, Ref. [30] had argued that the number of fundamental frequencies supporting a drive constitutes a notion of “drive complexity”: intuitively, with (fewer) more frequencies, trajectories through Hilbert space are (less) more complicated and thus (less) more ergodic. Indeed, this is rigorized by no-go theorems [30], which preclude k -HSE if there are too few tones, in line with our finding that the Floquet drive (which has a single fundamental frequency) fails to be ergodic at any level.

However, the smoothly kicked QP and Fibonacci drives display vastly different levels of ergodicity, yet are both two-frequency QP drives, indicating that the number of fundamental frequencies is not a complete governor of ergodicity. We observe a key difference lies in the *regularity* of their kick modulation vectors, Eqs. (3) and (4): $\vec{g}(\phi)$ for the former (latter) is a smooth (step) function, resulting in kicks whose amplitudes are smoothly (discontinuously) modulated. Although seemingly technical, we speculate these lead to physical consequences:

regularity of drives is directly related to their underlying *distribution* of frequencies, i.e., their power spectra, with smoothly (discontinuously) modulated drives being more (less) localized in energy; see the End Matter for plots. This results in fewer (more) transition channels between different levels in the system, such that one may expect less (more) Hilbert space exploration (see past works in the quantum chaos literature, which discussed similar effects [58,59]). Formalizing this observation, as well as incorporating physical constraints like symmetries and conservation laws, would be important next steps in achieving a more complete theory of HSE, the formulation of which would amount to a conceptual advancement of our understanding of nonequilibrium quantum statistical mechanics.

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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End Matter

Power spectra of smoothly-kicked QP and Fibonacci drives—Here, we compute the power spectra of the smoothly kicked QP and Fibonacci drives. The power spectrum of a time-dependent Hamiltonian quantifies how the operator’s strength is distributed over frequencies. It is defined as follows. Let $H(t)$ be a Hamiltonian defined on integer times $t \in \mathbb{N}$ (we assume discrete times to match the Hamiltonians we implement in this Letter; the definition for the power spectrum of Hamiltonians for continuous time follows analogously). Its Fourier component is $\tilde{H}(\omega(k)) = \sum_{t=0}^{T-1} H(t)e^{-i\omega(k)t}$, where $\omega(k) = 2\pi k/T$ and $k = 0, \dots, T-1$. Here, T is a large, finite-time cutoff (related to how long the Hamiltonian is applied for). Then $H(t)$ ’s power spectrum is defined to be

$$P(\omega(k)) = \frac{1}{T^2} \|\tilde{H}(\omega(k))\|^2, \quad (\text{A1})$$

where $\|\cdot\|$ is the operator norm.

In Fig. 4 we plot the power spectra of the smoothly kicked QP drive and the Fibonacci drive implemented in the experiments. We see that the power spectrum of the smoothly kicked QP drive is highly localized in frequency

space, being concentrated on the frequencies $\omega = 0, 2\pi - 2\omega_2, 2\omega_2$. In contrast, the power spectrum of the Fibonacci drive is delocalized over frequency space. These features arise from the *regularity* of the drives: the former is a drive whose kick amplitudes are modulated smoothly, while the latter is a drive whose kick amplitudes are modulated discontinuously.

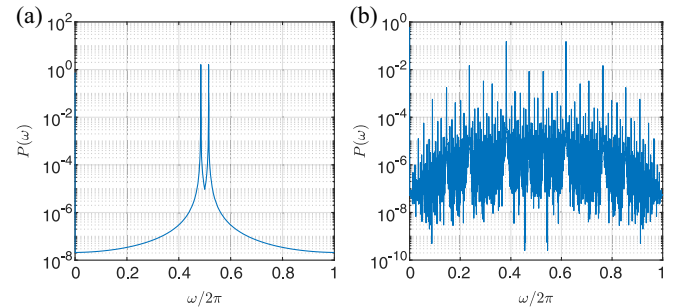


FIG. 4. Power spectra of driven Hamiltonians. (a) Power spectrum of the smoothly kicked QP drive, defined by Eqs. (1) and (3). (b) Power spectrum of the Fibonacci drive, defined by Eqs. (1) and (4). In these plots, we took $T = 1000$.