Critical prethermal discrete time crystal created by two-frequency driving

Discrete time crystals are non-equilibrium many-body phases of matter characterized by spontaneously broken discrete time-translation symmetry under periodic driving. At sufficiently high driving frequencies, the system enters the Floquet prethermalization regime, in which the periodically driven many-body state has a lifetime vastly exceeding the intrinsic decay time of the system. Here, we report the observation of long-lived prethermal discrete time-crystalline order in a three-dimensional (3D) lattice of $^{13}$C nuclei in diamond at room temperature. We demonstrate a two-frequency driving protocol, involving an interleaved application of slow and fast drives that simultaneously prethermalize the spins with an emergent quasi-conserved magnetization along the $\hat{x}$ axis. This enables continuous and highly resolved observation of their dynamic evolution. We obtain videos of the time-crystalline response with a clarity and throughput orders of magnitude greater than previous experiments. Parametric control over the drive frequencies allows us to reach time-crystal lifetimes of up to 396 Floquet cycles, which we measure in a single-shot experiment. Such rapid measurement enables detailed characterization of the entire phase diagram, highlighting the role of prethermalization in stabilizing the time-crystal response. The two-frequency drive approach expands the toolkit for investigating non-equilibrium phases of matter stabilized by emergent quasi-conservation laws.
harnesses prethermalization. This has also inspired extensions of time-crystal phenomena to classical systems^{26–28}. Nevertheless, characterizing the full phase diagram of the emergent prethermal DTC order and rigidity, and elucidating its thermalization dynamics towards infinite temperature, remains a challenging task^{29–31}. At the same time, this is critical for advancing our fundamental understanding of non-equilibrium order and for leveraging such collective phenomena in applications, such as in quantum simulation and sensing.

In this Article we report on a novel experimental approach for the high-throughput characterization of the formation and melting of PDTCs, which permits measuring their phase diagram with unprecedented high resolution. Our experiments are performed on an interacting system of driven, hyperpolarized, $^{13}$C nuclear spins in diamond (Fig. 1a), endowed with long prethermal lifetimes under Floquet driving. We propose a novel use of a two-frequency Floquet drive (note that the term ‘frequency’ refers to the inverse periods of the two superimposed drives, rather than their Fourier decompositions) applied to the $^{13}$C nuclei (Fig. 1b): the spins are made to prethermalize to an effective Hamiltonian $\mathcal{H}_c$ (featuring a quasi-conserved $\mathbf{x}$-magnetization; Supplementary Information), under a fast drive (with period $\tau$), while also being periodically kicked away from the prethermal state by a slower drive with period $T = N\tau$. The deviation away from the $\mathbf{x}$ axis can be continuously monitored in periods between the fast drive, allowing a means to track the full system dynamics for long periods (>14 s), corresponding to 450 Floquet cycles or $\sim 1.35 \times 10^5$ fast pulses, without collapsing the quantum state. Compared to the point-by-point measurements of previous experiments, this allows a dimension reduction for mapping the PDTC phase diagram. In concert with the multiple-minute-long $T$ lifetimes of the $^{13}$C nuclei, we are able to unravel the emergent prethermal order with substantially higher clarity than previous measurements. The developed two-frequency drive represents a generalization of conventional Floquet driving, allowing for a mutable design and the experimental realization of non-equilibrium order even beyond the paradigmatic DTC.

**System**

We consider a lattice of $^{13}$C nuclei (Fig. 1a) in diamond, optically hyperpolarized for $t_{\text{pol}} = 60$ s by surrounding nitrogen vacancy (NV) centres^{32–34}. Hyperpolarization yields an ~680-fold enhancement in $^{13}$C magnetization over thermal equilibrium, yielding a starting density matrix $\rho_0 \sim \mathcal{E}$, with the $z$-polarization fraction $\mathcal{E} = 0.68\%$ (Methods). Here, $\mathcal{E} = \sum_{j \neq 0} c_j$, with $c_j$ being a Pauli operator associated with nuclear spin $j$ (ref. 32). The natural abundance ($1\%$) of $^{13}$C nuclei are not spatially ordered, and are coupled via dipolar interactions, $\mathcal{H}_d = \sum_{k \neq \ell} c_k c_\ell$. One can define a characteristic energy scale via the median interspin coupling, $J = 0.66$ kHz (ref. 33). They are also subject to electron-mediated random on-site fields from surrounding NV and Pt paramagnetic defects, $\mathcal{E}_c = \sum_{k} c_k$ (refs. 34, 35).

Despite the concomitant position disorder and random on-site fields, the 3D long-range nature of the interactions precludes many-body localization. The long-range interactions also make simulating the exact Floquet dynamics for a large number of spins inaccessible using classical computers.

Figure 1b describes the experimental protocol: hyperpolarized $^{13}$C nuclei are tipped along $\mathbf{x}$, and subject to concatenated ‘fast’ and ‘slow’ Floquet drives, characterized by periods $\tau$ and $T = N\tau$, respectively. The fast drive, consisting of a train of $\mathbf{\theta}(\pi)$ kicks (Fig. 1b), engineers the internuclear Hamiltonian so that spins, initially aligned along $\mathbf{x}$, are rendered quasi-stationary$^{27}$. This is accomplished by arranging $\mathcal{H}_{\text{rad}} \rightarrow \mathcal{H}_c + \delta\mathcal{E}_{\text{c}} \gamma_L$ to leading order in the Magnus expansion, such that $[\mathcal{H}_c + \delta\mathcal{E}_{\text{c}} \gamma_L, J_{k\ell}^{\gamma_L}] = 0$ (Supplementary Section VI). This quasi-conservation causes the spins to prethermalize along $\mathbf{x}$ due to the non-integrable character of $\mathcal{H}_c$. By contrast, in the absence of the fast drive, evolution under $\mathcal{H}_{\text{rad}}$ causes system observables to rapidly (in $\tau^{-1}$) become indistinguishable from a featureless infinite temperature state.

Interspersed at period $T$, the slower drive kicks the spins along the $\mathbf{y}$ (or $\mathbf{z}$) axis with angle $\gamma$ (Fig. 1b). The spins are allowed to prethermalize back along $\mathbf{x}$ between successive kicks (Fig. 2a). Figure 1c shows this visually on the Bloch sphere (in the rotating frame) for a kick of angle $\gamma = \pi + \epsilon$. Therefore, in the prethermal plateau, the system is governed by an effective Hamiltonian obtained through an inverse frequency expansion (Supplementary Section VI). For $\gamma = 0, \pi$, although the slow $\gamma$ kicks do not cause any extra heating, they give rise to non-equilibrium ordered states. At $\gamma = \pi$, the two-cycle time-evolution operator $U_\gamma^{(2)} = \exp[-i\gamma\mathcal{H}_c]$ is governed by the $x_\gamma$ symmetric many-body Hamiltonian $\mathcal{H}_c$, where the $x_\gamma$ symmetry is implemented by flipping the $x$ direction of all spins. This drive-induced symmetry of $U_\gamma^{(2)}$ together with the discrete time-translation invariance, creates a spatio-temporal eigenstate order in $U_\gamma^{(2)}$. Any initial state that breaks this symmetry is forced to oscillate with period $2\gamma$, forming a PDTC state. In the experiment, we additionally observe that interactions stabilize a finite region near $\gamma = \pi$, where a stable PDTC period-doubling response arises, with the spins flipping from $+\mathbf{x}$ to $-\mathbf{x}$ between successive kicks (Fig. 1c).

A distinguishing feature of our experiments is the ability to quasi-continuously track the prethermalization dynamics after each kick. The spins are non-destructively interrogated by nuclear magnetic resonance (NMR) in $t_{\text{pol}} = 0.67$ s windows between the fast pulses (Fig. 1b). The magnitude and phase of the $^{13}$C Larmor precession is sampled every 1 ns, allowing one to reconstruct the instantaneous projections $\mathcal{P}(\gamma, t)$, as well as the phase $\phi(t)$ of the spin vector in the $\mathbf{x} - \mathbf{y}$ plane (Methods). In reality, the quasi-continuous time variable $t$ is discretized in units of $\tau$. Hyperpolarization enables high signal-to-noise (SNR $\geq 10^3$) signal acquisition per measurement point.
Prethermalization and discrete time crystals

It is worth emphasizing that, although rotating-frame DTCs are also observable in our system under a suitable single-frequency drive, either no continuous measurement can be performed or the spins rapidly decay to infinite temperature near the DTC point \( \gamma = \pi \) (Supplementary Section VI). Two-frequency driving circumvents this problem, because \( \theta \) can be arbitrarily chosen (except for \( \theta = \{0, \pi\} \)) and DTCs instead appear conditioned on \( \gamma \). Concatenated driving thus engineers a separation between the interaction-driven spin dynamics and breaking of a discrete spatio-temporal symmetry (Supplementary Section VII). Additionally, the ratio \( N = T/\gamma \) is tunable, affording flexibility in exploring dynamical regimes at small and large \( N \). In typical experiments, \( NM > 1.35 \times 10^5 \) fast pulses are applied, corresponding to \( M > 450 \) Floquet cycles. Our approach portends observing the long-time and intra-period time-crystalline dynamics continuously and without state re-initialization. This constitutes a vast improvement in throughput with respect to contemporary experiments (Supplementary Videos 1–4), with a resulting measurement speedup of at least \( NM > 10^3 \); compared to experiments probing lab-frame (2) DTCs\(^2\), these gains could be as much as \( NM(T_f/\tau_{\text{pad}}) \approx 10^7 \).

**Prethermalization and discrete time crystals**

Figure 2a first clarifies the prethermalization process during the spin kicks, essential to ultimately generate the PDTCs. Shown is a single-shot trace plotting \( \langle J_x \rangle \) after every fast pulse. We display the Floquet cycle number \( M \) on the lower \( x \) axis and absolute time on the upper \( x \) axis for an exemplary 165-ms window. Here, \( \theta = \pi/2 \), and the \( y \) kicks are applied every 50.7 ms (denoted by the dashed lines). Each \( y \) kick is associated with transient dynamics of the coupled \(^{13}\text{C}\) nuclei as they prethermalize along \( \hat{x} \), producing the flat plateau-like regions shown. The inset in Fig. 2a zooms into one representative transient. The oscillation period here is set approximately by the number of fast pulses required to complete a \( 2\pi \) rotation. The high temporal resolution reveals by the high SNR, and the ability of the \(^{13}\text{C}\) nuclear system to sustain a large number of \( y \) kicks (also Fig. 3a) allow us to track the kicked prethermalization dynamics for long periods.

Employing an exemplary choice of flip angle \( \gamma = 0.97\pi \) slightly away from the perfect DTC point, Fig. 2b demonstrates the generation of stable DTC order, exhibiting period-doubling in \( \langle J_x(t) \rangle \) during the application of \( M > 450 \) \( y \) kicks (Fig. 3a presents the full data). The data were collected in a single run of the experiment. Red (blue) colours here represent odd (even) \( y \) kicks, and prethermal plateaux separate successive \( y \) kicks. These data comprise more than \( 1.35 \times 10^5 \) fast pulses, and rigid DTC behaviour is observable for \( >14 \) s (also Fig. 3a). The decay of the signal is approximately mono-exponential with a \( 1/e \) lifetime of \( t = 4.68 \) s (corresponding to \( M = 149 \) Floquet cycles at \( \gamma = 0.97\pi \)), making it among the longest-lived DTCs observed in the literature. Moreover, the \( J_f \) state value here is considerably beyond the state of the art for systems exhibiting DTC order\(^{4,12,13,19}\), demonstrating an ability to probe long-time dynamics in our system. This long-time stability can be attributed to the emergent quasi-conservation of \( x \) magnetization...
under the evolution engineered by the two-frequency drive. In particular, our driving protocol allows for the formation and observation of DTC order, even if the temperature in the prethermal plateau is infinite, but also at room temperature, which may be well above the critical temperature associated with symmetry breaking (Supplementary Section VII). The long-range nature of the spin–spin interaction suggests critical DTC order. On the other hand, the lifetime of the DTC order is parametrically controlled by the frequency (of switching) of the employed drives (see the section Melting of prethermal order and Supplementary Section VII). The observed DTC order thus corresponds to a critical prethermal DTC.

Continuous observation yields an insightful view into the thermalization dynamics away from the stable points, a challenging task in other experimental systems. This is demonstrated in data focusing on the instantaneous phase \( \varphi(t) = \tan^{-1}(\langle J_x \rangle / \langle J_y \rangle) \) of the spins in the \( x - y \) plane after every fast pulse. This is captured by the points in Fig. 2c for the full 14-s experiment, and considering four representative constant-\( y \) values (Fig. 3a presents the full data). When \( y = 0.01\pi \), the spins are locked at \( \varphi = 0 \), reflecting prethermalization along \( \hat{x} \). A slight deviation, \( y = 0.06\pi \), reveals transient oscillations in \( \varphi \) with every kick but no sign inversion. The transients result in the observed data spreading around \( \varphi = 0 \). Figure 2c(ii) therefore permits visualization of the ‘melting’ of the prethermal \( \hat{x} \)-magnetization order to infinite temperature, where \( \varphi \) becomes random; this is observable at \( t \gtrsim 10 \) s. Analogously, the PDTC order (Fig. 2c(iii)), here at \( y = 1.01\pi \), is visible as characteristic \( \pi \)-phase switching between successive kicks. The last panel (Fig. 2c(iv)) denotes \( y = 1.08\pi \), when again PDTC melting can be observed via the phase randomization at long times.

Collating 285 such data traces while varying angle \( y \in (-1.1\pi, 1.1\pi) \), it is possible to construct a video of the kicked prethermalizing spins (see ancillary .gif files in the Supplementary Information). The result is plotted in Fig. 3a, where we display \( \langle J_z \rangle \) (colourbar), with each vertical slice corresponding to measurement data as in Fig. 2b. The data highlight almost three decades in the slow drive kicks, for which we observe PDTC order, indicated by a regular switching of the magnetization from \( \hat{x} \) to \( -\hat{x} \) with every slow kick (here 300 fast pulses). The transition into

**Fig. 3** Prethermal DTC phase diagram. 
(a) Emergence of prethermal DTCs. A total of 285 traces similar to Fig. 2b are plotted stacked for different values of kick angle \( y \in [-1.1\pi, 1.1\pi] \). Colours represent signal \( \langle J_z \rangle \) (colourbar). Time \( M \) and Floquet cycle number \( M \) run vertically and are plotted on a logarithmic scale. Data are taken to 14 s (\( M = 450 \) cycles). The central feature (near \( y = 0 \)) shows stabilization of long-time spin survival as a result of drive-induced quasiconservation of \( \hat{x} \)-magnetization due to Floquet prethermalization. The PDTC response (striped regions) is visible near \( y = \pm \pi \). The striped signal denotes spins flipping between \( \hat{x} \) and \( -\hat{x} \) in a period-doubled fashion. Spins flip every \( N \) fast pulses, and the rigid PDTC response (peaks) persists to three decades of the fast drive. The remaining regions are characterized by rapid spin decay due to dipolar interactions, and are indistinguishable from the infinite temperature state. Insets (i) and (ii): zoom into the PDTC response in two 1-s windows centred at \( t = 3.15 \) s and \( t = 11.55 \) s, respectively, plotted on a linear scale. Rigid DTC behaviour is observable for over 14 s, even at \( y = 0.97\pi \). See ref. 38 for the video version of the same dataset. (b) Fourier transform of the data in (a), plotted with respect to the inverse period of the slow drive \( \omega = 2\pi/14 \) rad. Colours represent the strength of the Fourier peak intensity spanning six orders of magnitude (colourbar). PDTC (period-doubled) order is evident from the extended white peaks at \( (y, \omega) = (\pm \pi, \pi) \) and the trivial (prethermal) phase arises for \( (y, \omega) = (0, 0) \). Dashed lines indicate the expected Fourier peak pattern in the absence of interactions. (c, d) Numerical simulations analogous to the experiments in (a) and (b), respectively, for \( L = 14 \) interacting spins on a pseudo-random graph with \( J_{\tau} = 0.07 \) and \( N = 300 \) (Supplementary Section IV). There is excellent qualitative agreement with experimental observations.
and out of the finite PDTC regions is clearly evident, and not easily accessible in other experiments, allowing us to precisely characterize the heating dynamics (Fig. 4). Interestingly, we observe very similar heating dynamics in the prethermal regime around \(\gamma = 0\) and the PDTC regime around \(\gamma = \pi\), even though both regimes are associated with completely different non-equilibrium order.

Insets (i) and (ii) in Fig. 3a present zooms into PDTC regions in a 1.05-s window at \(t = 3.13\) s and \(t = 11.55\) s, respectively. Although faint, the stable periodic DTC response is clearly evident, even in Fig. 3a(ii), highlighting the high SNR in the experiment.

A complementary view of Fig. 3a is presented in Fig. 3b, where we consider the Fourier transform of \(\langle \mathcal{J}_y(t) \rangle\), the mean signal value between successive \(y\) kicks for each value of \(y\) (Fig. 3a, vertical slices). Plotted is the corresponding Fourier intensity on a logarithmic scale, where a span to six orders of magnitude is visible. The PDTC response appears as a sharp period-doubling peak in frequency at \((y, \omega) = (\pm \pi, \pi)\). Using a 20% magnitude threshold, we estimate the finite \(y\)-extent of the rigid PDTC regions to \(\Delta y \approx \pm 0.2\pi\) about \(y = \pm \pi\). Similarly, the long-lived prethermal phase at \(y = 0\) appears as a sharp peak at \((y, \omega) = (0, 0)\). For comparison, the dark regions correspond to rapid state decay near \(y = \pm \pi/2\), where the effective Hamiltonian no longer features a quasi-conserved magnetization (Supplementary Section VI). The dashed lines in Fig. 3b indicate the expected position of the Fourier peaks in the absence of interactions, \(J_0 = 0\). The difference in the experimental data is evident, which indicates the role played by interactions in our system.

Comparing the experimental measurements to corresponding numerical simulations (Supplementary Sections V and VII–IX), we find excellent qualitative agreement: in Fig. 3c,d we display the exact simulation results matching the experimental conditions in the data in Fig. 3a,b. Interestingly, we observe that the long-range interactions, together with the random spin positions, induce a self-averaging effect in the simulations so that reliable theoretical results can already be obtained for moderately small system sizes (here \(L = 14\) spins). By contrast, the experimental platform comprises a cluster of about \(L = 10^3\) interacting spins\(^{35}\), which outcompetes numerically reachable system sizes by three orders of magnitude. The presence of a large number of interacting degrees of freedom is crucial for experimentally observing collective statistical mechanics phenomena such as symmetry breaking and thermalization. Thus, our results indicate that Floquet-engineered \(^{13}\)C nuclei can serve as a competitive quantum simulator of thermalizing spin dynamics.

The observed PDTC behaviour is also insensitive to the initial state. Although we lack microscopic control over the initial state, different and highly non-trivial initial states can be obtained from letting the initial hyperpolarized density matrix \(\rho_0\) evolve under the system Hamiltonian \(\mathcal{H}_{\text{dd}} + \mathcal{H}_z\) up to times \(\tau\). For a set of such states we find comparable results between experiment and simulation (Supplementary Section VII). Our system thus satisfies all required landmarks of PDTCs: a parametrically long-lived prethermal window featuring spatio-temporal symmetry breaking, which is rigid over a finite \(y\) region, and insensitivity to fine-tuned initial states.

### Melting of prethermal order

To quantify the stability of the PDTC order away from the stable point at \(y = \pi\), we investigate the influence of finite \(\epsilon\) on its lifetime, which we define as the \(1/e\) decay time of the signal. In turn, the inverse lifetime defines the associated heating rate.

Note that, even at \(y = \pi\) (where the slow drive does not contribute to heating), the system slowly heats up due to energy absorption resulting from the fast (spin-locking) \(x\) drive. This provides a lower bound for the relevant heating rates, \(\Gamma_{\text{min}}(\tau)\), which we find scales as a power law of the \(x\)-drive period, \(\Gamma_{\text{min}} \propto (\Gamma)^{-1}\) (ref. 33). Finite \(\epsilon\) opens an additional channel for the system to absorb energy, and, eventually, the heating rates of both drives conspire to yield a combined overall heating rate \(\Gamma(\tau, \epsilon)\). In Fig. 4 we extract the heating rates associated to finite \(\epsilon\), and we display \(\Gamma - \Gamma_{\text{min}}\) away from the stable points \((y = 0, \pi)\) obtained from Fig. 3a,b. The curves indicate a parametrically controllable power-law heating with an exponent of \(-2.21\), consistent with Fermi’s golden rule. Intriguingly, we observe that the power-law heating rates are close to identical for both stable points \((y = 0, \pi)\), where the relevant timescales are governed by a Lorentzian \(\Gamma^{-1} \propto (\Gamma^2 + \Gamma_{\text{min}}^2)^{-1}\) for some system-dependent constant \(c\); this behaviour is also borne out in numerical simulations (Supplementary Section VIII and Supplementary Fig. 14).

For the observed prethermal DTC lifetime to be controllable, the relevant lifetimes are expected to increase with increasing drive frequencies. Indeed, the heating time in units of Floquet cycles \(\Gamma^{-1}/N\) depends sensitively on the frequency of the Floquet drive \(\omega_\nu = 2\pi \nu/\tau\). In particular, at \(\epsilon = 0\), our model predicts \(\Gamma^{-1}/N \approx 1/(N\nu^2)\). Increasing the frequency of switching by tuning \(N(\tau)\) is expected to lead to a linear (quadratic) increase of \(\Gamma^{-1}/N\), which we have confirmed numerically for our system (Supplementary Section VII and Supplementary Fig. 15). In Fig. 4c,d we display experimental measurements of the dependence of \(\Gamma^{-1}/N\) as a function of \(N\) and \(\tau\), for finite \(\epsilon\). Although a crisp \(1/N\) dependence is washed out in the presence of finite \(\epsilon\), we observe a clear increase in the lifetime. For sufficiently large \(N\) the timescale-separated regime \(\Gamma/\nu \gg 1\), \(\Gamma(t)/N\) as a function of \(t\) agrees well with a power law with an exponent close to \(-2\), whereas at very small \(\tau\), the DTC lifetime increases to a halt. This is to be expected for fixed \(\epsilon\), because decreasing the value of \(\tau\) reduces the many-body nature of the effective Hamiltonian (Supplementary Section VI). Note that DTC order is a many-body
effect that relies on spontaneous spatio-temporal symmetry breaking in interacting systems. Thus, when the many-body nature is gradually reduced, the stability and rigidity of the DTC order are expected to decline accordingly.

### Two-frequency Floquet engineering

When the condition \( N = T/\tau \gg 1 \) is not met, timescale separation between the slow and fast drives is violated, and a complex interplay between the two drives emerges, leading to novel effective Hamiltonians that sensitively depend on \( N \). Consequently, the corresponding heating diagrams exhibit substantial differences, as compared to the timescale-separated case, but also for different values of \( N \). In Fig. 5 we show the time evolution of the \( x \) magnetization as a function of \( \gamma \) for \( N = 8 \) and \( N = 9 \). Three features are particularly noteworthy: (1) in contrast to the timescale-separated case, the heating dynamics around \( \gamma = \pi \) differs substantially from that around \( \gamma = 0 \). In particular, (2) far away from \( \gamma = \pi \), heating depends sensitively on the specific value of \( N \), where even a minimal change (in \( N \)) can induce completely different heating behaviours (compare Fig. 5a,b). These features can be explained by the different effective Hamiltonians (Supplementary Section IX) forming at different \( N \), \( \gamma \), respectively. (3) Even though interference effects induce new effective Hamiltonians with case-specific heating properties, the formation of time-crystalline order is stable against these deformations (Fig. 5c,d). However, the relevant lifetimes and regimes of rigidity depend sensitively on \( N \) (Supplementary Section IX): changing \( N = 8 \) into \( N = 9 \) amounts to a late lifetime increase from 270 to 396 Floquet cycles (cf. Fig. 5c,d). These results provide a proof-of-principle example of the fine interplay between the two drives, which offers a versatile tool to engineer new kinds of effective Hamiltonians with orchestrated physical properties, such as DTC order and beyond.

### Conclusions and outlook

Summarizing, we have observed critical prethermal discrete time crystals in dipolar coupled nuclear spins in a bulk 3D solid. We have developed a novel protocol to excite and observe PDTC formation and melting using a concatenated two-frequency Floquet drive. Parametric control over both drive frequencies allows us to reach PDTC lifetimes up to 396 Floquet cycles, observable in a single run of the experiment. This experimental advance unveils properties of the PDTC with a high degree of clarity, including its rigidity and melting characteristics. Our measurements are in excellent agreement with numerical simulations, and approximate theoretical predictions.

Our study greatly expands the Floquet engineering toolkit: it portends multi-frequency concatenated drives to excite and stabilize novel types of quantum matter far from equilibrium by engineering emergent quasi-conservation laws that offer protection against immediate high-temperature melting.

An extension of this work to multi-frequency driving is well within the scope of present-day experimental capability, and can be used to introduce more degrees of freedom to Floquet engineering. We envision application of these ideas in quantum simulation and sensing in atomic, molecular and optical (AMO) platforms as well as with hyperpolarized prethermal spins in solid-state systems.

### Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01891-7.

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38. Video of full dataset from Fig. 2b and Fig. 3a of main text (first 55 Floquet cycles) https://youtu.be/m5iASnBZ9oo (2021).
Methods

Sample and hyperpolarization methodology
We used a chemical vapour deposition-grown single crystal of diamond with a defect density of 1-ppm NV centres and natural abundance $^{13}$C. The sample was placed with its [100] face parallel to the hyperpolarization and readout magnetic fields (38 mT and 7 T, respectively). $^{13}$C nuclei were hyperpolarized via the NV centres via a protocol described in ref. 33 involving continuous optical pumping and swept microwave irradiation. Polarization was transferred from the NV electrons to proximal $^{13}$C nuclei via a spin-ratchet mechanism that involves traversals of Landau–Zener anticrossings in the rotating frame of the swept microwaves. The traversal probabilities are nuclear spin-state selective and are biased towards one nuclear spin orientation; this results in hyperpolarization. Spin diffusion serves to transfer this polarization to nuclear spins in the bulk lattice. The typical bulk polarization levels reached are ~0.6%.

Data collection and processing
The data-processing pipeline follows a similar approach to that described in ref. 33. The NMR signal was sampled continuously in $t_{\text{acq}}$ windows between the fast spin-lock pulses, at a sampling rate of 1 Gsamples s$^{-1}$ via a Tabor Proteus arbitrary waveform transceiver. Continuous observation exploits the fact that the NMR coil produces no backaction on the spins. In typical experiments, the pulses were spaced apart by $\tau = 105$ μs and the acquisition windows were $t_{\text{acq}} = 64$ μs. The $^{13}$C Larmor precession (at ~73 MHz) was heterodyned to 20 MHz before digitization. For each $t_{\text{acq}}$ acquisition window, we took a Fourier transform and extracted the magnitude and phase of the 20-MHz peak. This corresponds to application of a digital bandpass filter with a linewidth of $f_{\text{pass}} = 31.2$ kHz. Fast digitization hence yields SNR gains, and the typical SNR per point (detection window) is $\gtrsim 10^2$. For the 14-s-long acquisition in this Article, there were ~135,000 such data-collection windows.

Extraction of amplitude and phase in Figs. 2 and 3
The magnitude and phase of the Fourier transform of the heterodyned precession in each $t_{\text{acq}}$ readout window report, respectively, on the magnitude and phase of the spin vector in the $x \rightarrow y$ plane in the laboratory frame. This corresponds to magnitude $S_x = |\langle \hat{S}_x \rangle| + |\langle \hat{S}_y \rangle|$ and phase $\phi_x = \langle \hat{S}_y \rangle / \langle \hat{S}_x \rangle$, where subscript L refers to the laboratory frame. It is more convenient to instead obtain the phase of the spins $\phi \approx \phi$ in the rotating frame. To do this, we note that the phase values $\phi_x$ obtained in successive $t_{\text{acq}}$ windows just differ by the (trivial) phase accrued during the $t_{\text{acq}}$ spin-locking pulse. Subtracting this global phase allows us to extract $\phi$, which in combination with the magnitude signal then allows us to extract the survival probability along the $x$ direction in the rotating frame, $\langle \hat{S}_x(t) \rangle$, that we display in Figs. 2 and 3.

Numerical simulations
To numerically simulate the many-body dynamics of dipolar interacting $^{13}$C nuclei, we designed random graphs of $L = 14$ and $L = 16$ interacting spins-1/2 and performed exact time evolution with up to $10^9$ kicks (corresponding to $\sim 10^7$ y kicks) based on an open multi-processing (OMP)-parallelized Krylov method using the open-source Python package QuSpin$^{39}$. For further details we refer the reader to Supplementary Section IV.

Comparison with previous work
DTC order has been studied in a range of systems, from cold atoms$^{12,29}$ over superconducting qubits$^{32}$ to systems based on NV centres$^{11,16}$. The DTC order observed in these works can be separated into two groups: many-body localized DTCs and prethermal DTCs. Although many-body localized DTCs are assumed to be infinitely long-lived in the absence of decoherence, prethermal DTCs, as in our experiments, are ultimately limited by the lifetime of the prethermal plateau. However, in reality, many-body localized DTCs are also subject to decoherence due to technical limitations. Remarkably, even though our DTC order is of a different physical origin, we find comparable lifetimes to state-of-the-art many-body-localized DTCs. Moreover, our lifetimes exceed those reported for prethermal DTCs, both in units of Floquet cycles as well as in absolute time.

Apart from these benchmark parameters, our system shares ingredients with the systems investigated in refs. 11 and 16. As in our work, these works investigate systems based on NV centres: ref. 16 examines a quasi-1D system of nine dipolar coupled $^{13}$C nuclear spins in the many-body localized regime, whereas in ref. 11, effective two-level systems of electronic states in NV centres are used to implement DTC order. In contrast, our (3D) system consists of $10^3$–$10^4$ dipolar coupled $^{13}$C nuclear spins. In particular, in comparison to ref. 11, the normalized interaction strength $|J_1/J_2|$ in our system is increased by a factor $4.5 \times 10^3$ (here $\gamma_{en}$ denotes the gyromagnetic ratio of electronic/nuclear spins); moreover, at comparable normalized driving strength, we obtain an improvement of normalized spin lifetimes by a factor of $5.7 \times 10^2$ (see also Supplementary Section III for a detailed table of relevant system parameters). Induced by the two-frequency drive, the DTC order we observe is prethermal; that is, our DTC lifetime is parametrically controlled by the drive frequency, whereas no such feature is present in ref. 11 where a single-frequency drive is used.

Data availability
Data from experiments and simulations displayed in the main text are available in Zenodo with the identifier https://doi.org/10.5281/zenodo.7301638. All other data from the Supplementary Information are available from the authors upon reasonable request.

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Author contributions
W.B., C.F., M.B. and A. Ajoy conceived the research. W.B., A.P., E.d.L.S., A. Akkiraju, J.D.A., S.C., P.R., E.D. and A. Ajoy set up the experimental apparatus, performed measurements and analysed the data. C.F. performed the numerical simulations and the perturbative analysis. A. Ajoy and M.B. supervised the experiment and the theory work.

Competing interests
The authors declare no competing interests.
Additional information
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