Quasi-Floquet Prethermalization in a Disordered Dipolar Spin Ensemble in Diamond

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Floquet (periodic) driving has recently emerged as a powerful technique for engineering quantum systems and realizing nonequilibrium phases of matter. A central challenge to stabilizing quantum phenomena in such systems is the need to prevent energy absorption from the driving field. Fortunately, when the frequency of the drive is significantly larger than the local energy scales of the many-body system, energy absorption is suppressed. The existence of this so-called prethermal regime depends sensitively on the range of interactions and the presence of multiple driving frequencies. Here, we report the observation of Floquet prethermalization in a strongly interacting dipolar spin ensemble in diamond, where the angular dependence of the dipolar coupling helps to mitigate the long-ranged nature of the interaction. Moreover, we extend our experimental observation to quasi-Floquet drives with *multiple* incommensurate frequencies. In contrast to a *single*-frequency drive, we find that the existence of prethermalization is extremely sensitive to the smoothness of the applied field. Our results open the door to stabilizing and characterizing nonequilibrium phenomena in quasiperiodically driven systems.

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Floquet theory describes the dynamics of a system whose Hamiltonian exhibits a *single* time-translation symmetry. Often used as a tool to control quantum systems, Floquet engineering (i.e., periodic driving) can help to prevent environment-induced decoherence and, more recently, has enabled the study of novel quantum dynamical phenomena [1–42]. In particular, many-body Floquet systems can host intrinsically nonequilibrium phases of matter, ranging from discrete time crystals [6,7,10–12,18,19,43,44] to Floquet topological states [15,20,22,45–51]. Even richer nonequilibrium behaviors can arise in "quasi-Floquet" systems, where a *single* time-translation symmetry is replaced by *multiple* time-translation symmetries [21,35,52–55]. For instance, the spontaneous breaking of the latter can result in time quasicrystalline order, which features a subharmonic response that is fundamentally distinct from conventional time crystals [21,35].

A critical obstacle to stabilizing and observing such phenomena in driven quantum systems is Floquet heating: the inevitable absorption of energy from the driving field. One potential solution arises when the driving frequency, ω, is significantly larger than the local energy scale, J, of the many-body system; in this case, Floquet prethermalization occurs and there exists an exponentially long-lived preheating regime described by an effective time-independent Hamiltonian, $\mathcal{H}_{\mathrm{eff}}$ [23–30]. The intuition underlying Floquet prethermalization is simple—in order to absorb a single photon from the drive, the system must undergo $\sim\omega/J$ off-resonant rearrangements of its local degrees of freedom. This higher-order process leads to an exponentially slow heating rate $\sim\mathcal{O}(e^{-\omega/J})$.

Despite this promise, there are two natural scenarios where prethermalization can break down: (i) systems with long-range, power-law interactions [14,33] and (ii) quasi-Floquet systems where multiphoton processes can enable resonant energy absorption [35].

Here, we report the experimental observation of Floquet prethermalization in a long-range interacting quantum system under quasi-periodic driving. Our experimental platform consists of a dense ensemble of dipolar interacting nitrogen-vacancy (NV) centers in diamond (Fig. 1) [7,8,40,56–58]. With single-frequency modulation, we observe that the heating time, τ^*, is consistent with an exponential scaling with increasing driving frequency. In contrast, by driving quasi-periodically with two frequencies, we find that Floquet heating can be fitted to a stretched exponential profile with $\tau^* \sim \mathcal{O}(e^{\omega^{(1/2)}})$ [35]. Interestingly, in the quasiperiodic case, the heating is extremely sensitive to the *smoothness* of the drive; indeed, when the system is driven via rectangular pulses (as opposed to sinusoidal pulses), we observe a significant enhancement in the heating rate (Fig. 3) [35]. We remark that the presence of slow heating is reminiscent of classic results from the NMR literature detailing the observation

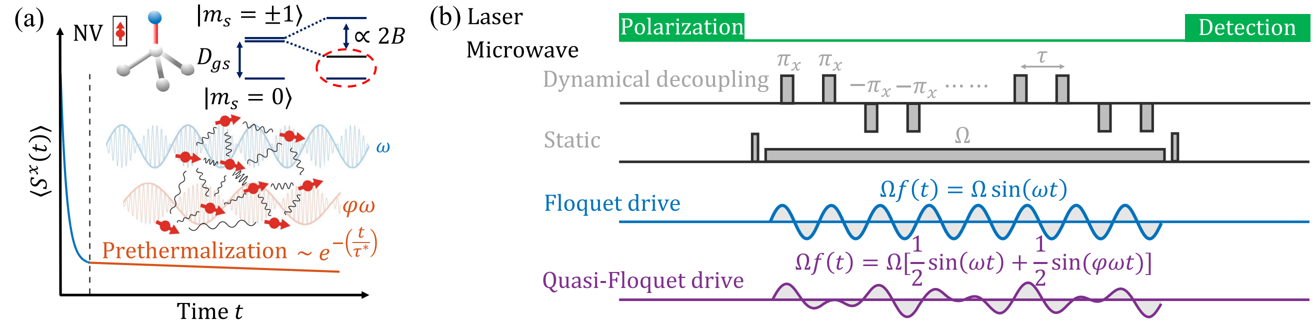

FIG. 1. Quasi-Floquet prethermalization in a dipolar spin system. (a) Quasiperiodic driving of a strongly interacting NV ensemble in diamond with two incommensurate frequencies ω and $\varphi\omega$, where φ is the golden ratio. Typical thermalization dynamics of the spin polarization $\langle S^x(t)\rangle$ exhibit an initial fast decay followed by a late-time slow relaxation. Top: Level structure of the NV center. Without an external field, $|m_s = \pm 1\rangle$ sublevels are degenerate and sit $D_{gs} = (2\pi) \times 2.87$ GHz above $|m_s = 0\rangle$. A magnetic field $B \sim 350$ G along the NV axis splits $|m_s = \pm 1\rangle$, enabling the isolation of a two-level system. (b) Experimental sequence. The dynamical decoupling sequence eliminates the on-site random fields induced by the environmental bath spins. The sequence includes a series of fast π pulses with alternating phases along $\hat{x}$ and $-\hat{x}$ axes to compensate the pulse errors. The interpulse spacing is fixed at $\tau = 0.1$ μs, much smaller than the interaction timescale between NV centers. A static microwave $\Omega \sum_i S_i^x$ together with dipolar interaction serves as the static Hamiltonian $\mathcal{H}_0$, and a time-dependent microwave $\Omega f(t) \sum_i S_i^x$ serves as the Floquet and quasi-Floquet drives. A final $\pi/2$ pulse along the $\mp\hat{y}$ axis rotates the spins back to $\hat{z}$ for detection [64].

of long-lived dynamics in driven systems [38,59–63]. However, the origin of the long-lived dynamics is either expected to be independent of the driving frequency or exhibits a lifetime that scales as a power law of the driving frequency [64]. These scenarios are markedly distinct from the context of Floquet prethermalization, which exhibits an exponentially long lifetime in ω. Despite such distinction, how to unequivocally demonstrate exponential over power-law scaling from real experiment remains a challenging task [64].

Experimental system.—We choose to work with a diamond sample containing a dense ensemble of spin-1 NV centers with concentration, $\rho \sim 4.5$ ppm [56,64]. The NV centers can be optically initialized and read out using green laser. In the presence of an external magnetic field ~350 G, the $|m_s = \pm 1\rangle$ sublevels are Zeeman split, allowing us to isolate an effective two-level system, $\{|m_s = 0\rangle, |m_s = -1\rangle\}$ [Fig. 1(a)]. By applying a resonant microwave field with Rabi frequency Ω, the effective Hamiltonian governing the system (in the rotating frame) is [7,8,57]:

$$\mathcal{H}_0 = -\sum_{i<j} \frac{J_0 \mathcal{A}_{i,j}}{r_{i,j}^3} (S_i^z S_j^z - S_i^x S_j^x - S_i^y S_j^y) + \Omega \sum_i S_i^x, \quad (1)$$

where $J_0 = (2\pi) \times 52$ MHz $\cdot$ nm^3, $\mathcal{A}_{i,j}$ characterizes the angular dependence of the dipolar interaction, $r_{i,j}$ is the distance between the ith and jth NV centers, and $\hat{S}$ is spin operator.

We note that $\mathcal{H}_0$ contains only the energy-conserving terms of the dipolar interaction under the rotating-wave approximation. The approximation holds because the NV transition frequency is more than 3 orders of magnitude larger than any other terms in the interacting Hamiltonian [64]. The presence of other paramagnetic spins in the diamond lattice [77] leads to an additional on-site random field at each NV center that is eliminated using dynamical decoupling [Fig. 1(b)] [64].

Let us begin by characterizing the dynamics of the NV ensemble under the static Hamiltonian $\mathcal{H}_0$. We set $\Omega = (2\pi) \times 0.05$ MHz, comparable to the average dipolar interaction strength. After optically initializing the NV spins to $|m_s = 0\rangle$, we then prepare a product state, $\otimes_i [(|0\rangle_i + |-1\rangle_i)/\sqrt{2}]$, by applying a global $\pi/2$ pulse around the $\hat{y}$ axis. We let the system evolve under $\mathcal{H}_0$ for a time t, before measuring the final NV polarization, $\langle S^x(t)\rangle$, along the $\hat{x}$ direction.

The polarization dynamics proceed in two steps. At early times, $t \lesssim 100$ μs, the polarization exhibits rapid decay toward a plateau value, reflecting local equilibration under $\mathcal{H}_0$ [Fig. 2(a)]. Following these initial dynamics, the system exhibits a slow exponential decay $\sim A_0 e^{-(t/T_0)}$ with $A_0 = (0.43 \pm 0.01)$ and a timescale $T_0 = (0.82 \pm 0.03)$ ms that is consistent with spin-phonon relaxation [78]. To ensure that the observed spin dynamics does not come from the incorporated dynamical decoupling pulses, we also investigate the corresponding spin dynamics at $\Omega = 0$. The measured NV polarization quickly decays to zero, in agreement with the expectation of thermalization behavior [Fig. 2(a), inset].

Floquet prethermalization.—To probe the nature and existence of Floquet prethermalization, we modulate the Rabi frequency as $\Omega(t) = \Omega[1 + f(t)]$, where $f(t) = \sin(\omega t)$ [Fig. 1(b)]. We note that $\Omega(t)$ contains two fundamentally different components: the constant field, $\Omega \sum_i S_i^x$, is a part of the previous undriven Hamiltonian $\mathcal{H}_0$,

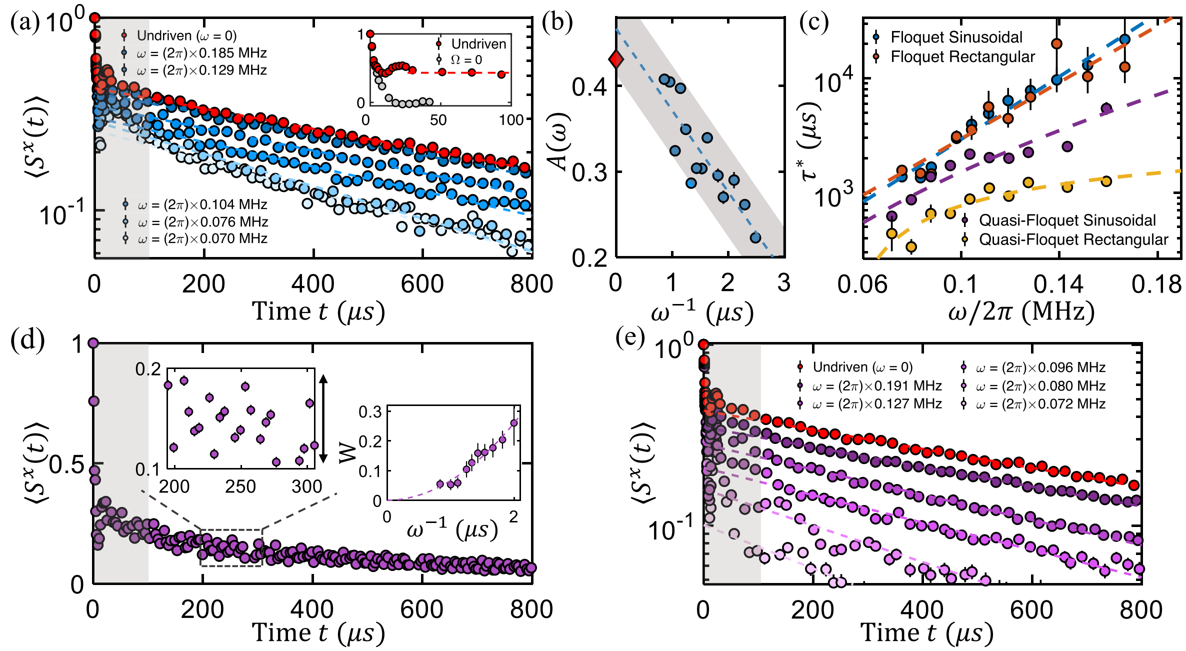

FIG. 2. Probing the spin dynamics under periodic and quasiperiodic drives. (a) The measured spin polarization $\langle S^x(t)\rangle$ under Floquet drive. After an initial fast decay (gray shaded area), a long-lived prethermal regime persists. Dashed lines are fits of the late-time dynamics using single exponential decay. Inset: Measured initial spin dynamics for the undriven case (red): After the initial fast relaxation, the polarization decays to an equilibration plateau (dashed line), due to the finite static field, $\Omega\sum_i S_i^x$. In contrast, the polarization with only dynamical decoupling ($\Omega = 0$) quickly decays to zero. (b) Measured prethermal equilibrium value, $A(\omega)$, as a function of ω^{-1}. The dashed line is a linear fit with the gray shaded area representing 95% confidence interval. The red diamond marks the measured amplitude for the undriven case. (c) Heating timescale τ^* as a function of the driving frequency ω. For both sinusoidal and rectangular single-frequency drives, $\tau^* \sim \mathcal{O}(e^{\omega/J})$. For sinusoidal quasiperiodic drive, $\tau^* \sim \mathcal{O}(e^{\omega^{(1/2)}})$; while for rectangular quasiperiodic drive, $\tau^* \sim \mathcal{O}(\omega^{(1/2)})$. (d) Measured spin dynamics under quasiperiodic drive [$\omega = (2\pi)\times 0.103$ MHz]. We observe an additional small time-quasiperiodic micromotion on top of a slow relaxation. Left inset: enlargement of the micromotion. Right inset: relative amplitude of the micromotion W scales quadratically with ω^{-1}. (e) Using the rolling average to remove the micromotion [64], we observe a quasi-Floquet prethermal regime, whose lifetime τ^* increases with ω. Dashed lines are fits using single exponential decay. Error bars on the spin polarization represent 1 s.d. accounting for statistical uncertainties, and error bars on the prethermal plateau and timescale represent 1 s.d. from the fitting.

while the time-dependent component $\Omega f(t)\sum_i S_i^x$ acts as the Floquet drive. Starting with a driving frequency $\omega = (2\pi)\times 0.07$ MHz, which is comparable to energy scales within $\mathcal{H}_0$, we perform the same spin polarization measurement [light blue curve in Fig. 2(a)]. The measured spin dynamics at stroboscopic times, $t = 2\pi N/\omega$ (where N is an integer), exhibit an initial relaxation, which is qualitatively similar to the undriven case. However, the late-time dynamics exhibit a significantly faster polarization decay, arising from Floquet heating. To obtain the heating timescale τ^*, we fit the experimental data to $\sim Ae^{-[(t/\tau^*)+(t/T_0)]}$, where T_0 is the previously extracted spin-phonon lifetime. As shown in Fig. 2(a), by increasing the driving frequency, one can extract the frequency dependence of the amplitude, $A(\omega)$, and the heating timescale, $\tau^*(\omega)$; both are crucial for understanding the nature of Floquet prethermalization.

Focusing first on the heating timescale, we find that τ^* increases exponentially with ω for more than an order of magnitude, demonstrating the existence of a parametrically long-lived prethermal regime [Fig. 2(c)]. The observed exponential scaling also allows us to extract a phenomenological local energy scale of the NV many-body system, $J_{\rm exp} = (2\pi)\times(0.032\pm 0.006)$ MHz.

Intuitively, this $J_{\rm exp}$ extracted from the Floquet heating process is expected to agree with the local energy scale of the system. However, for systems with power-law interaction as ours ($\sim 1/r^3$ in three dimensions), a naive estimation of the local energy scale $J \approx \int (J_0/r^3)\rho d^3\mathbf{r}$ is divergent, and thus, one should expect the prethermalization to not exist. Nevertheless, an important missing piece to this puzzle is the angular dependence of the dipolar interaction, $\mathcal{A}_{i,j}$ [33]. Crucially, the combination of this angular dependence and the NV's random positioning in the diamond lattice ensures that the average, $\bar{\mathcal{A}}_{i,j} = 0$, which helps to mitigate the divergence of the above integral [79].

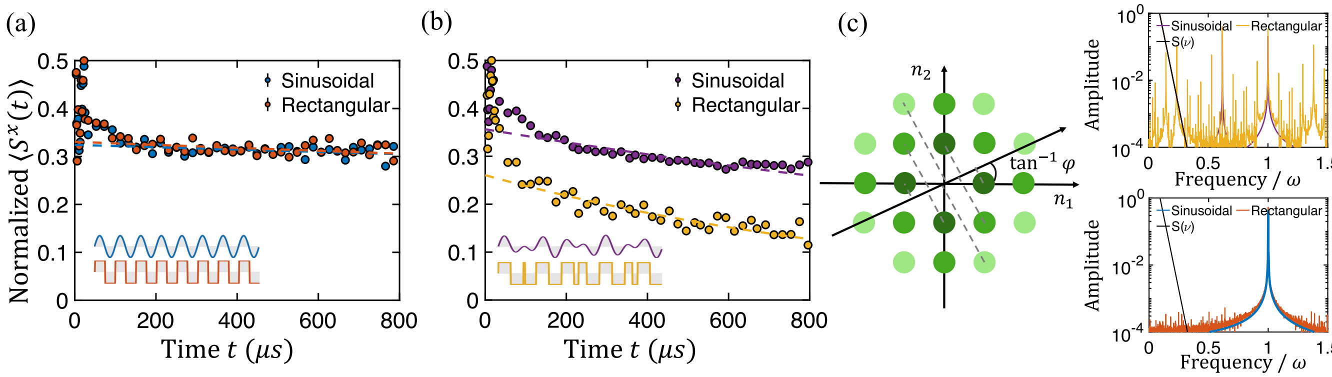

FIG. 3. Comparison between sinusoidal and rectangular driving fields. (a) A representative late-time dynamics of the spin ensemble under single-frequency (Floquet) drive [$\omega = (2\pi) \times 0.151$ MHz]. Here the plotted polarization is normalized by the intrinsic decay timescale, T_0, measured without the driving field, i.e., normalized $\langle S^x(t) \rangle \sim e^{-t/\tau^*}$. (b) Under quasiperiodic drive [$\omega = (2\pi) \times 0.143$ MHz], the spin dynamics is extremely sensitive to the smoothness of the drive, and displays a significantly faster decay under a rectangular drive. Error bars represent 1 s.d. accounting statistical uncertainties. (c) Two-dimensional Fourier spectrum F_{n_1,n_2} of the driving field $f(t)$ [64]. The horizontal (vertical) axis corresponds to the absorption of n_1 (n_2) photons with energy ω ($\varphi\omega$) from the drive, where $n_1, n_2 = 0, \pm 1, \ldots$. The color of each green dot represents the amplitude of F_{n_1,n_2} at frequency $\nu = n_1\omega + n_2\varphi\omega$ (darker represents larger amplitude). The Fourier spectrum $F(\nu)$ can be thought of as a projection of green dots onto a line with slope φ. Right panel: Fourier spectrum $F(\nu)$ of quasiperiodic and periodic drive with sinusoidal and rectangular amplitude. For the quasi-Floquet scenario, the spectrum with rectangular drive exhibits resonances at the small frequency range, which overlaps with the system's local spectral function $S(\nu)$ (black) and leads to energy absorption.

A more careful analysis reveals that the relevant local energy scale is the variance of the interaction, $\tilde{J} \approx [\int (J_0 \mathcal{A}_{i,j}/r^3)^2 \rho d^3\mathbf{r}]^{(1/2)} = \sqrt{(16\pi/15)} J_0 \rho$ [64]. As long as the driving frequency $\omega > \tilde{J}$, one should expect the presence of prethermalization, in agreement with previous theoretical studies [33]. Using the independently calibrated NV density, ρ, we estimate the local energy scale $\tilde{J} \approx (2\pi) \times 0.02$ MHz, which is indeed comparable with J_{exp} extracted from Floquet heating.

Next, let us turn to analyzing the frequency dependence of the amplitude, $A(\omega)$. One can think of $A(\omega)$ as the value of the prethermal plateau. In general, for short-range interactions, it is expected that $A(\omega)$ is determined by a time-independent effective Hamiltonian, $\mathcal{H}_{\text{eff}}(\omega) = \mathcal{H}_0 + \mathcal{O}(\tilde{J}/\omega)$, which can be calculated order-by-order via a Magnus expansion [80]. In this case, $A(\omega) = \text{Tr}[\sum_i S_i^x e^{-\beta H_{\text{eff}}(\omega)}] = A_0 + \mathcal{O}(\tilde{J}/\omega)$, where the inverse temperature β, is set by the energy density of the initial state.

For sufficiently long-range interactions (such as dipolar interactions in three dimensions), the existence of $\mathcal{H}_{\text{eff}}$ is unproven [14,26,80–82]. However, by probing the functional form of $A(\omega)$ and its extrapolated value as $\omega \to \infty$, one can provide experimental evidence for the existence of $\mathcal{H}_{\text{eff}}$. As depicted in Fig. 2(b), we find that the frequency dependence of $A(\omega)$ is linear in ω^{-1}, allowing us to extrapolate $A(\omega \to \infty) = (0.47 \pm 0.06)$. This is consistent with the measured value in the undriven case, $A_0 = (0.43 \pm 0.01)$, suggesting that despite the presence of strong long-range interactions, the effective Hamiltonian exists and can be well-approximated by $\mathcal{H}_0$ at leading order [64].

Quasi-Floquet prethermalization.—We now turn to the quasi-Floquet setting. Specifically, we choose $f(t) = \frac{1}{2}[\sin(\omega t) + \sin(\varphi\omega t)]$, where $\varphi = (\sqrt{5} - 1)/2 \approx 0.618$ is the golden ratio, so that the system is driven by two incommensurate frequencies. From the perspective of Floquet heating, the situation is significantly more complex. In particular, recall that within Fermi's golden rule, the heating rate can be estimated from the overlap between the Fourier spectrum of the drive, $F(\nu) = \int f(t) e^{i\nu t} dt$, and the local spectral function of the spin ensemble, $S(\nu) = \sum_{i,j} \delta(E_i - E_j - \nu) |\langle i | S^x | j \rangle|^2$, where E_i and $|i\rangle$ are the eigenenergy and eigenstate of the spin system.

This picture immediately provides a more formal intuition for the exponentially slow heating observed in the context of single-frequency driving [Fig. 3(c)]. In particular, for $f(t) = \sin(\omega t)$, $F(\nu)$ exhibits a cutoff at frequency ω. Meanwhile, as aforementioned, $S(\nu)$ exhibits an exponentially small tail for frequencies $\nu > \tilde{J}$ [83]. In combination, this implies that for a single driving frequency, $\omega > \tilde{J}$, energy absorption is strongly suppressed leading to $\tau^* \sim e^{\omega/\tilde{J}}$.

For driving with two incommensurate frequencies, even when $\omega > \tilde{J}$, there are multiphoton processes that are effectively resonant within the local energy scale; these processes correspond, for example, to the absorption of n_1 photons of energy ω and the emission of n_2 photons of energy $\varphi\omega$. Thus, there is no strict cutoff for $F(\nu)$, and the drive spectrum exhibits a nonzero amplitude for all frequencies $\nu = |n_1\omega - n_2\varphi\omega| < \tilde{J}$. Interestingly, despite this, for sufficiently large driving frequencies, seminal results have proven that the quasi-Floquet heating timescale remains extremely slow, exhibiting a stretched exponential lower bound [84].

In contrast to the Floquet case, we measure the dynamics at evenly spaced time points, since there does not exist a stroboscopic time which is an integer multiple of *both* drives. Much like the single-frequency drive, after an early-time transient, the spin polarization exhibits a slow decay. However, we observe small oscillations scaling as $\sim\omega^{-2}$ on top of the decay [Fig. 2(d)], corresponding to the micromotion of the quasi-Floquet system [85–87]. We note that for a single-frequency drive, similar micromotion will also emerge if one does not measure the spin dynamics at stroboscopic times [64]. Intuitively, such micromotion arises from the time-dependent portion of the Hamiltonian which only averages to zero for each complete Floquet cycle. To reliably extract a heating timescale τ^* from our quasi-Floquet measurements, we perform a rolling average to obtain the overall decay profile [Fig. 2(e)] [64]. By varying the driving frequency, we extract a heating timescale, $\tau^* \sim e^{\omega^{(1/2)}}$ [Fig. 2(c)], which is consistent with the theoretically predicted stretched exponential form [35].

Robustness of quasi-Floquet prethermalization.—The stability of slow prethermal heating is quite different depending on whether one considers the Floquet or quasi-Floquet setting. For the Floquet setting, the exponential behavior of τ^* is robust to the functional form of the drive amplitude $f(t)$. However, in the quasi-Floquet setting, the stretched exponential behavior of τ^* is *only* predicted to hold when $f(t)$ is smooth. In particular, when $f(t)$ is smooth, even though $F(\nu)$ does not exhibit a cutoff for small ν, its amplitude is exponentially small in this regime [Fig. 3(c)] [35,36].

These expectations are indeed borne out by the data [Figs. 2(c) and 3]. Using a rectangular wave $f(t) = \mathrm{Sgn}[\frac{1}{2}\sin(\omega t) + \frac{1}{2}\sin(\varphi\omega t)]$, we observe that the heating timescale is significantly shortened and scales as a power-law with increasing driving frequency $\sim\omega^{(1/2)}$, as opposed to a stretched exponential. In contrast, for a single-frequency drive, the smoothness of the driving field is not critical: the Floquet heating timescale exhibits an exponential scaling for both sinusoidal and rectangular forms of $f(t)$.

Outlook.—Looking forward, our Letter opens the door to a number of intriguing future directions. First, it is interesting to ask whether the restriction on $\bar{\mathcal{A}}_{i,j} = 0$ is essential for realizing prethermalization in long-range interacting systems [14,33,34]. Second, the observed long-lived quasi-Floquet prethermal regime can enable the experimental realization of novel nonequilibrium phases of matter [15,20,22,45–51]. Finally, while our experiments suggest the presence of power-law-slow heating in the case of a quasi-Floquet, rectangular-wave drive, the precise frequency dependence of the heating rate remains unknown and requires future study.

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