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Phys. Rev. Lett. **119**, 010602 — Published 6 July 2017

DOI: 10.1103/PhysRevLett.119.010602

Critical time crystals in dipolar systems

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(Dated: May 30, 2017)

We analyze quantum dynamics of periodically driven, disordered systems in the presence of long-range interactions. Focusing on stability of discrete time crystalline (DTC) order in such systems, we use a perturbative procedure to evaluate its lifetime. For 3D systems with dipolar interactions, we show that the corresponding decay is parametrically slow, implying that robust, long-lived DTC order can be obtained. We further predict a sharp crossover from the stable DTC regime into a regime where DTC order is lost, reminiscent of a phase transition. These results are in good agreement with the recent experiments utilizing a dense, dipolar spin ensemble in diamond [Nature 543, 221-225 (2017)]. They demonstrate the existence of a novel, *critical* DTC regime that is stabilized not by many-body localization but rather by slow, critical dynamics. Our analysis shows that DTC response can be used as a sensitive probe of non-equilibrium quantum matter.

Introduction.—Understanding and controlling non-equilibrium quantum matter is an exciting frontier of physical science. While periodic driving has long been used to control the properties of quantum systems, it was only recently realized that periodically driven (Floquet) systems can also host new states of matter that are not possible in equilibrium. In particular, this is possible in a class of driven disordered systems exhibiting many-body localization (MBL) [1], called Floquet-MBL systems, which avoid unbounded heating to infinite temperature [2–4]. The latter is generally expected to befall all ergodic isolated systems due to external driving [5–7].

One remarkable example of a novel, non-equilibrium phase of matter is the recently introduced discrete time crystal (DTC) [8–11], which is characterized by a spontaneously broken discrete time-translation symmetry of the underlying drive. In such systems, physical observables exhibit robust oscillations with a period that is an integer multiple of the underlying driving period T. Indeed, key signatures of such robust DTC order have been observed in two recent experiments [12, 13]. In particular, one of these realizations involved a disordered ensemble of $\sim 10^6$ spins associated with nitrogen-vacancy (NV) centers in diamond, which interact between themselves via dipolar couplings [13]. The origin of apparent robustness of the observed DTC order in such a system [13], however, has not been fully understood. Although this system is disordered due to the random positions of the NV-centers in 3D, the long-range dipolar interactions are believed to preclude localization [14–18]. Moreover, a prethermal regime of the DTC [19], was also ruled out [13] since in the experiment the initial polarized state is effectively at an infinite temperature with respect to the effective Hamiltonian due to the randomly varying signs of the dipolar interactions. Since neither localization nor prethermalization are likely the mechanisms that stabilize the DTC order, this raises important questions about the origin of the observed robust DTC response.

This Letter develops a theoretical treatment of DTC order in systems with long-range interactions. We utilize a perturbative approach to analyze the interplay of long-range interactions, periodic driving, and positional disorder of spins. Focusing on dipolar systems in 3D, we show that although DTC order is only transient, it can persist for asymptotically long times with strongly suppressed thermalization rate. This behavior is intrinsically connected to slow thermalization dynamics of disordered dipolar systems in 3D, which has been previously shown to be consistent with the so-called critical regime [14, 20] without periodic drive. As a function of experimental parameters, we find that the relaxation time shows a sharp crossover between a regime where DTC response is robust and a regime where it decays rapidly. This crossover is reminiscent of a phase transition, thereby allowing us to obtain the effective phase diagram of DTC which is in good agreement with experimental results. Thus, our work provides an explanation of the recent experimental observations [13], and also demonstrates the possibility of the DTC in systems with critical dynamics, a regime which we refer to as 'critical time crystals'. Furthermore, our perturbative approach can be used to study the non-equilibrium properties in other driven disordered systems with long-range interactions.

Our key results can be understood by considering a simple spin model that describes an ensemble of dipolar interacting NV centers, used in the experiments of Ref. [13]. Using strong microwave excitations, the effective Ising interactions between spins were engineered, described by the following Hamiltonian:

$$H_0 = \sum_i \Omega S_i^x + \sum_{i,j} \frac{J_{ij}}{r_{ij}^3} S_i^x S_j^x, \tag{1}$$

where $\vec{S}_i = (S_i^x, S_i^y, S_i^z)$ are Pauli spin-1/2 operators, Ω the strong microwave driving along $\hat{x}$, J_{ij} the orientation dependent coefficient of dipolar interactions with typical strength J_0, and r_{ij} the distance between spins i and

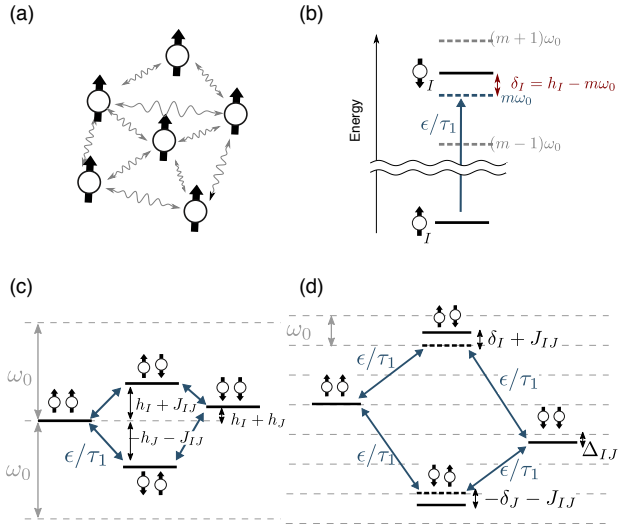

Figure 1. (a) Ensemble of randomly positioned spins in 3D interacting via dipolar interactions. (b) Illustration of single-spin-flip processes. (c, d) Energy level diagram for the second-order process of two spins flipping, in two regimes: (c) high frequencies ($\omega_0 \gg W$) and (d) low ($\omega_0 \ll W$) frequencies. The applied field flips a spin with magnitude ϵ/τ_1, which costs energy $\sim h_I - m^*\omega_0$.

j. We assume that the spin-1/2 particles are randomly distributed in three dimensional space with density n_0 and neglect coupling to the environment [Fig. 1(a)]. DTC order was observed by interrupting the evolution under Hamiltonian (1) with rapid, global pulses that rotate the spin ensemble along the $\hat{y}$-axis by an angle $\pi + \epsilon$. The corresponding Floquet unitary is given by

$$U_F = \exp\left[-i\sum_i (\pi+\epsilon) S_i^y\right] \exp\left[-iH_0\tau_1\right], \qquad (2)$$

where τ_1 is the period for which the spins are allowed to interact for. In the experiment, the period is chosen such that $\Omega\tau_1 = 2\pi n$, and therefore Ω can effectively be taken to be 0 in Eq. (1). When the system is initialized in a state where all spins are polarized along the $+\hat{x}$ direction, a non-trivial temporal response may be revealed by measuring the average polarization $P(n\tau_1)$ of the ensemble along $\hat{x}$ after n Floquet cycles, or equivalently, $q(n) \equiv (-1)^n P(n\tau_1)$, which serves as an order parameter for the DTC phase. The stability of the DTC order can be ascertained by studying the decay rate of $q(n)$ for large number of cycles as a function of τ_1 and ϵ.

In order to describe the dynamics of $q(n)$, we move into a so-called toggling frame, which rotates by $P_\pi \equiv \prod_j \exp\left[-i\pi S_j^y\right]$ each time a global pulse is applied to the system. Since $P_\pi S_i^x (P_\pi)^{-1} = -S_i^x$, the $2\tau_1$-periodic oscillation in $P(n\tau_1)$ naturally appears as a time-indepdent spin polarization in this new frame. The dynamics of the system is then described by the Floquet unitary $\bar{U}_F = \exp\left[-i\sum_i \epsilon S_i^y\right] \exp\left[-iH_0\tau_1\right]$, or, equivalently, by an effective time-dependent Hamiltonian

$$H(t) = \sum_{ij} \frac{J_{ij}}{r_{ij}^3} S_i^x S_j^x + \epsilon \sum_i S_i^y \sum_n \delta(t - n^-\tau_1). \qquad (3)$$

Thus, our problem reduces to studying the depolarization dynamics of an initialized polarized spin ensemble under the time evolution of $H(t)$.

Physical picture.—The essence of our analysis is to study resonant spin dynamics that lead to depolarization perturbatively in ϵ, while accounting for energy exchanges provided by the external drive. In particular, since $\sum_n \delta(t - n^-\tau_1) = \frac{1}{\tau_1}\sum_m e^{im\omega_0 t}$, the pulsed periodic spin rotations can be viewed as spin excitation with harmonics of the fundamental frequency $\omega_0 \equiv 2\pi/\tau_1$ and fixed magnitude ϵ/τ_1. While this driving allows energy absorption and emission in integer multiples of ω_0, the interplay of strong interactions and positional disorder suppresses direct energy exchanges such that typical spins depolarize only via indirect higher order processes in ϵ.

Let us first consider the case without perturbations, i.e. $\epsilon = 0$. Then the polarization of each spin along $\hat{x}$ is conserved. When all spins are initially polarized, each spin therefore experiences a mean-field potential $h_i \equiv \sum_{j\neq i}(J_{ij}/r_{ij}^3)\langle S_j^x\rangle$. Due to the random positioning of spins, the strength of h_i is also random with zero mean and variance $W^2 = \langle \frac{1}{4}(\sum_{j\neq i} J_{ij}/r_{ij}^3)^2\rangle$, where $\langle\cdot\rangle$ denotes averaging over different positions.

When $\epsilon \neq 0$, there is depolarization due to spins experiencing a time-varying on-site field along the $\hat{y}$-axis. Let us therefore consider the first order process where spins individually flip due to the action of this field. If a spin experiences a a strong mean-field potential h_i compared to the applied field, that is, if $h_i\tau_1 \gg \epsilon$, then it does not flip – it experiences an effective field that is approximately pointing along the $\hat{x}$-axis and therefore precesses around it without significant depolarization. On the other hand, if $h_i\tau_1$ is close to an integer multiple of 2π, then the spin rotates along the $\hat{y}$-axis and depolarizes. Physically, this corresponds to an effectively resonant excitation of (individual) spins that arises when one of the driving harmonics is tuned close to their energy: $|h_i - m^*\omega_0| < \epsilon/\tau_1$ for some optimal integer m^* [see Fig. 1(b)]. Such resonances occur with a small probability in the limit of $\epsilon \ll W\tau_1$, and amount to a reduction of the total polarization by a constant factor proportional to $\epsilon/(\min(W,\omega_0)\tau_1)$. However, if $\epsilon \sim W\tau_1$, a substantial fraction of spins rapidly depolarize due to resonant processes shown in Fig. 1(b). Note that the phenomenological phase boundary extracted in Ref. [13], based on the existence of self-consistent closed spin trajectories, is consistent with the perturbative condition $\epsilon \ll W\tau_1$.

We next focus on the second order process illustrated in Fig. 1(c,d) in which a pair of spins I and J simultaneously flip their polarizations while exchanging their energies with each other *and* with the external drive. Such

processes are resonant when

$$\Delta_{IJ} \approx |h_I + h_J - m^* \omega_0| < J_{IJ}^{\text{eff}}, \tag{4}$$

where h_I and h_J are effective on-site potential for spins I and J, m^* is the optimal harmonic number that minimizes the energy difference, and J_{IJ}^{eff} is the effective amplitude of the pair-flip process. The amplitude J_{IJ}^{eff}

$$J_{IJ}^{\text{eff}} \sim \left(\frac{\epsilon}{\tau_1}\right)^2 \frac{J_{IJ}}{r_{IJ}^3} \left(\frac{1}{\delta_I^2} + \frac{1}{\delta_J^2}\right), \tag{5}$$

can be estimated from the interference of two paths in second order perturbation theory, as illustrated in Fig.1(c,d). Here $\delta_{I(J)} \equiv \min_{\ell^*}(h_{I(J)} - \ell^* \omega_0)$ is the energy difference between initial/final states and intermediate virtual states, up to extra energy provided by a driving harmonic ℓ^*. We find that J_{IJ}^{eff} is an effective long-range interaction decaying as $\sim 1/r_{IJ}^3$ allowing the flipping of remote spin pairs.

The resonance condition (4) is sensitive to ω_0 and behaves qualitatively differently in two limiting cases: (i) $\omega_0 \gg W$ and (ii) $\omega_0 \ll W$ [see Fig. 1(c,d)]. In the former case, the optimal choice is $m^* = \ell^* = 0$ since spins cannot absorb or emit such a large energy ω_0. In the latter case, effective energy differences (both Δ_{IJ} and δ_I, δ_J) are bounded by ω_0 as the external drive can always compensate energy in units of ω_0. These considerations yield the scaling $J^{\text{eff}}(r) \sim C J_0 / r^3$ with

$$C \approx \begin{cases} (\epsilon/\tau_1 W)^2 & \text{for } \omega_0 \gg W \\ \epsilon^2 & \text{for } \omega_0 \ll W \end{cases}, \tag{6}$$

and the effective range W^{eff} of the energy differences Δ_{IJ} becomes $W^{\text{eff}} \sim W$ for $\omega_0 \gg W$ and $W^{\text{eff}} \sim \omega_0$ for $\omega_0 \ll W$.

We now estimate the probability that a given spin finds a resonant partner within a ball of radius R. This is obtained by integrating the probability of finding such a partner in a shell R and $R + dR$

$$dP = \left(J^{\text{eff}}(R)/W^{\text{eff}}\right) n_0 4\pi R^2 dR, \tag{7}$$

from a short distance cut-off a_0 to R, which gives $P(R) \sim \log(R/a_0)$. Here the first factor in Eq. (7) is the probability of satisfying Eq. (4), and the second factor is the average number of spins within a shell of size R with the density n_0. As this probability diverges, it implies that pair-wise spin flips prevail, and the system thermalizes, with the DTC order slowly decaying over time. We can extract the timescale associated with these pair-spin flip processes using the typical distances R_* of resonant spin pairs. Solving $P(R_*) \sim 1$ gives $R_* \approx a \exp\left[W^{\text{eff}}/4\pi C J_0 n_0\right]$. Finally, the effective depolarization rate is estimated from the interaction strengths of typical pairs, i.e., $\tilde{\Gamma} \sim J^{\text{eff}}(R_*)$, leading to the decay

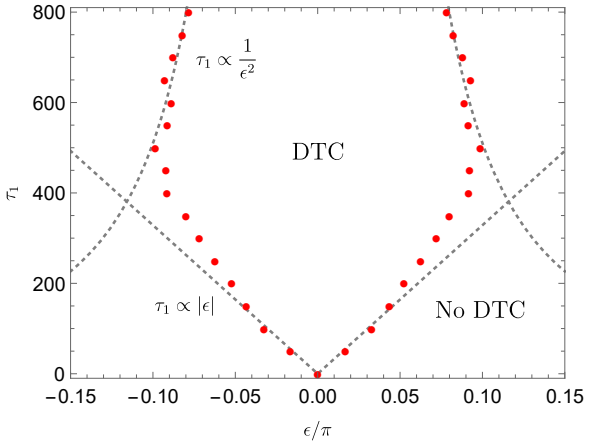

Figure 2. Phase diagram of the DTC obtained numerically (see [23] for details). Dotted lines indicate limiting behaviors of the phase boundary: at high driving frequencies, the phase boundary is linear, $\tau_1 \propto |\epsilon|$, while for low driving frequencies, it closes up as $\tau_1 \propto 1/\epsilon^2$, c.f. Eq. (8). This is in good agreement with the experimental observations of Ref. [13].

rate per Floquet cycle $\Gamma \equiv \tilde{\Gamma} \tau_1$:

$$\Gamma \sim \begin{cases} \frac{J_0 \epsilon^2}{a_0^3 \tau_1 W^2} \exp\left[-\frac{3 W^3 \tau_1^2}{4\pi J_0 n_0 \epsilon^2}\right] & \text{for } \omega_0 \gg W \\ \frac{J_0 \epsilon^2 \tau_1}{a_0^3} \exp\left[-\frac{3}{2 J_0 n_0 \epsilon^2 \tau_1}\right] & \text{for } \omega_0 \ll W. \end{cases} \tag{8}$$

This exponentially slow in $1/\epsilon^2$ decay of the DTC order is a central result of the present Letter, and is a direct consequence of critically slow thermalization of dipolar systems in 3D [20, 21]. Interestingly, the depolarization is exponentially sensitive to the parameters τ_1 and ϵ in two distinct ways: in regime (i) Γ is a function of τ_1^2/ϵ^2 while in regime (ii) it only depends on $1/\epsilon^2 \tau_1$. These considerations allow us to identify an effective phase boundary using the criteria $\tau_1^2/\epsilon^2 = A$ or $1/\epsilon^2 \tau_1 = B$ with some constants A and B. Remarkably, this boundary illustrated in Fig. 2 captures the key features observed in the experiment [13]: the linear growth of ϵ for short τ_1 and slow diminishing of ϵ at longer τ_1 [22].

Technical procedure.—We now outline the technical procedure that formalizes the above discussion (see [23] for details). The key idea is to identify a time-dependent unitary transformation of the Hamiltonian $H(t)$ such that non-resonant single spin-flips are essentially "integrated out" and only residual two-spin-flip processes become dominant terms in the effective Hamiltonian $H'(t)$. More specifically, we start from the Hamiltonian (3) with H_0 representing the Ising interactions and V the applied field, and perform a time-periodic unitary transformation $Q(t + \tau_1) = Q(t)$, which gives rise to:

$$H'(t) = Q(t)^\dagger (H_0 + \epsilon V \sum_n \delta(t - n^- \tau_1) - i\partial_t) Q(t). \tag{9}$$

Our goal is to eliminate terms that are linear in ϵ from $H'(t)$. Following Ref. [4], we look for $Q(t)$ of

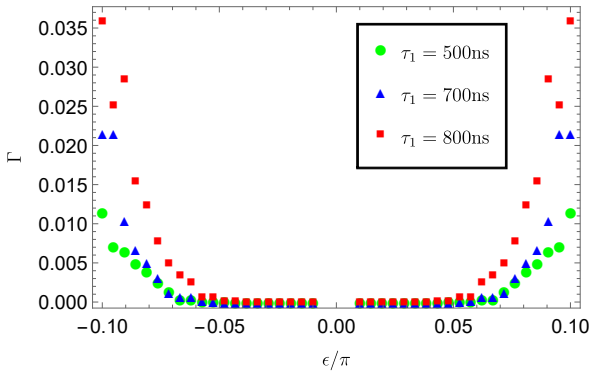

Figure 3. Decay rate versus perturbation ϵ for various τ_1s obtained numerically [23] . One sees a sharp rise of the decay rate as one crosses the DTC phase boundary (determined as the ϵ for which $\Gamma(\epsilon, \tau_1) = 1/100$), which is reminiscent of a phase transition.

the form $Q(t) = e^{\epsilon\Omega(t)}$ with anti-Hermitian operator $\Omega(t) = \sum_n \Omega^{(n)} e^{in\omega_0 t}$. Expanding Eq. (9) in powers of ϵ, and requiring that the $O(\epsilon)$ term equals 0 gives an equation for the nth Fourier mode $\Omega^{(n)}$:

$$\frac{V}{\tau_1} - [\Omega^{(n)}, H_0] + n\omega_0 \Omega^{(n)} = 0. \tag{10}$$

The matrix elements of the operator $\hat{\Omega}^{(n)}$ can be computed in the eigenstate basis $|s\rangle$ of H_0 (which is a product state basis in S_i^x operators):

$$\langle s'|\Omega^{(n)}|s\rangle = \frac{\langle s'|V|s\rangle}{(E_s - E_{s'} - n\omega_0)\tau_1}. \tag{11}$$

Noting that $V = \sum_i S_i^y$, the operator $\Omega^{(n)}$ has non-zero matrix elements only between spin configurations s and s' that differ by one spin-flip. If $|s\rangle$ and $|s'\rangle$ differ by the value of spin I, $E_s - E_{s'} = 2\sum_{j\neq I} \frac{J_{jI}}{r_{jI}^3} S_j^x(s) S_I^x(s) = 2h_I S_I^x$. We assume that the on-site field h_I is random (due to positional disorder and orientation dependence of J_{ij}) and sufficiently strong such that resonances are rare, i.e. the denominator in (11) typically does not diverge and the procedure controlled. Then, the rotated Hamiltonian to second order becomes

$$H'(t) = H_0 - \frac{\epsilon^2}{2}[\Omega(t), V] \sum_n \delta(t - n^- \tau_1) \tag{12}$$

A straightforward calculation [23, 24] using expression (11) gives an effective Hamiltonian of the following form:

$$H'(t) = H_0 + \sum_{IJ} \frac{A_{IJ} J_{IJ}}{r_{IJ}^3} \left(S_I^+ S_J^+ + h.c.\right) \sum_n \delta(t - n^- \tau_1), \tag{13}$$

where $S_I^+ \equiv (S_I^z + iS_I^y)/\sqrt{2}$ is the spin raising operator in S_I^x basis for the spin I, and A_{IJ} is the coefficient

$$A_{IJ} \approx -2S_I^x(s) S_J^x(s) \left(\frac{\epsilon}{\tau_1}\right)^2 \left(\frac{1}{\tilde{\delta}_I^2} + \frac{1}{\tilde{\delta}_J^2}\right), \tag{14}$$

where we introduced the notation $\frac{1}{\tilde{\delta}_J^2} = \sum_\ell \frac{1}{(h_J - \ell\omega_0)^2}$.

The effective Hamiltonian (13) contains the larger disordered part H_0, and long-range terms which can flip pairs of spins; the latter are suppressed proportional to ϵ^2, leading to slow relaxation. From Eq. (14) it is evident that the amplitudes for flipping a pair of spins depend on h_I, h_J, which in turn are determined by the positions of the spins. Assuming that h_I, h_J take typical values of the order W, and taking the contribution of the harmonic ℓ^* for which $h_J - \ell\omega_0$ is minimized (this gives the leading contribution to $\tilde{\delta}_J$), the expression (14) for the two-spin-flip amplitude reduces to the estimate (5) above.

We emphasize that the above unitary transformation is distinct from the rotating frame transformations employed to derive effective Hamiltonians in the high-frequency limit [25, 26]. Rather, it utilizes the randomness of our Hamiltonian in order to effectively integrate out non-resonant single spin flip processes.

Phase diagram.—Using the effective Hamiltonian approach described above, we obtain the phase diagram of the critical DTC. To improve upon the estimates for $\Gamma(\epsilon, \tau_1)$, we take into account the fact that the distribution of the potential h_i stems from positional randomness of spins, and numerically sample h_i from a distribution of 2000 spins in a 3D region with density 9.26×10^{-3} nm^{-3} with a short distance cut-off of 3 nm [20].

While Eq. (8) already provides analytical predictions for the decay rate Γ by estimating the typical distance R_* of resonant spin pairs, in numerics we find it more amenable to estimate Γ from an explicit depolarization in time profile; the counting arguments in Eq. (7) predicts a power-law decay of polarization $q(n)$, from which the decay timescale $1/\Gamma$ is extracted by equating $q(n)$ to a small threshold [23]. The phase boundary is then identified from a criteria $\Gamma(\epsilon, \tau_1) = \Gamma_* = 1/100$.

This approach yields the phase diagram illustrated in Fig. 2, which is in very good agreement with the experimental observations [13, 27]. At high driving frequency, the boundary approximately follows a relation $\tau_1 \propto |\epsilon|$ (also obtainable using a semi-classical argument), while at low frequency $\tau_1 \propto 1/\epsilon^2$, which indicates that DTC order becomes less stable as τ_1 is increased, due to the fact that multi-photon processes lead to faster depolarization. The DTC phase is most robust in the crossover regime, where $\omega_0 \sim W$.

We also note that strictly speaking DTC order has finite relaxation rate at any $\epsilon \neq 0, \tau_1 \neq 0$. However, we find that the relaxation rate Γ increases very sharply at a certain value of ϵ, as illustrated in Fig. 3, which matches the experimental observations and is reminiscent of a phase transition. Note, however, that unlike for a true phase transition, this increase does not become infinitely sharp even in the thermodynamic limit.

Summary and discussion.—We described a new approach to analyze dynamics of periodically driven spin

systems with long-ranged interactions and applied it to explain the recently observed surprising stability of DTC in dipolar spin system. The results of our analysis are in very good agreement with experimental observations. They demonstrate that these observations correspond to a novel, critical regime of the DTC order.

Furthermore, our general approach can be applied to analyze the interplay of long-range interactions, randomness, and periodic driving in a broad class of experimental systems. The present analysis focused on the experimentally relevant case of critical interactions, decaying as $1/r^\alpha$, where α coincides with the dimensionality of the system, $\alpha = d = 3$. This leads to direct relaxation processes of spin pairs. It is interesting to extend the analysis to the case $\alpha > d$ (e.g. $\alpha = 3, d = 2$), where resonant spin-pair-flip processes are rare and presumably do not provide the main relaxation channel. Experimentally, such a situation can be realized by reducing the dimensionality of the dipolar spins systems. In the static case, relaxation is expected to occur via multi-spin processes: in essence, a sparse resonant network may form, which can act as a heat bath that mediates relaxation of other spins [16, 18]. We expect that future experiments on DTC in reduced dimensions will allow one to probe such delicate interplay of various relaxation mechanisms in driven systems with long-range interactions. Our theoretical approach is well suited for analyzing such systems. Finally, apart from these specific realizations, our analysis demonstrates that the DTC response to periodic perturbations can be used as a sensitive probe of non-equilibrium quantum states and phases of matter.

We thank I. Protopopov, V. Khemani, J. Choi, R. Landig, H. Zhou, A. Vishwanath and N. Yao for useful discussions. This work was supported by Swiss National Science Foundation (W.W.H. and D.A.A), NSF, CUA, Vannever Bush Fellowship, ARO MURI, and Moore Foundation (S.C. and M.D.L.), and in part by the NSF under Grant No. NSF PHY11-25915 (W.W.H.,M.D.L., and D.A.A.). M.D.L., W.W.H. and D.A.A. are grateful to KITP, where this work was started, for hospitality during the program *Synthetic Quantum Matter*.

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strictly linear but will contain a logarithmic correction: $\tau_1 \approx c_1|\epsilon|\sqrt{\log(c_2/\Gamma_*) + \log|\epsilon|}$, valid for $|\epsilon| \gg \Gamma_*/c_2$. The constants c_1, c_2 can be easily obtained by equating Γ in Eq. (8) to $\Gamma_* = 1/100$ and solving, and are functions of the physical parameters J_0, n_0, W.

[23] Supplemental material online.

[24] Note that we have neglected flip-flop terms $S_I^+ S_J^- + h.c.$ since they are inconsequential for studying dynamics beginning from the initially polarized state in the $\hat{x}$ direction. Moreover, they do not give rise to depolarization as these terms are polarization conserving. See [23] for more discussion.

[25] D. Abanin, W. De Roeck, F. Huveneers, and W. W. Ho, "A rigorous theory of many-body prethermalization for periodically driven and closed quantum systems," ArXiv e-prints (2015), arXiv:1509.05386 [math-ph].

[26] Dmitry A. Abanin, Wojciech De Roeck, Wen Wei Ho, and François Huveneers, "Effective hamiltonians, prethermalization, and slow energy absorption in periodically driven many-body systems," Phys. Rev. B **95**, 014112 (2017).

[27] Note that this phase boundary is consistent with the perturbative condition $\epsilon \ll W\tau_1$.