Dissipative discrete time crystals

James O’Sullivan,1 Oliver Lunt,1 Christoph W. Zollitsch,1 M. L. W. Thewalt,2 John J. L. Morton,1 and Arijeet Pal1,3,4

1London Centre for Nanotechnology, University College London, 17-19 Gordon Street, London, WC1H 0AH, UK
2Department of Physics, Simon Fraser University, Burnaby, British Columbia, Canada
3Department of Physics, University College London, Gower Street, London, WC1E 6BT
4Rudolf Peierls Centre for Theoretical Physics, Oxford University, Oxford OX1 3PU UK
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Periodically driven quantum systems host a range of non-equilibrium phenomena which are unrealizable at equilibrium. Discrete time-translational symmetry in a periodically driven many-body system can be spontaneously broken to form a discrete time crystal, a putative quantum phase of matter. We present the observation of discrete time crystalline order in a driven system of paramagnetic P spin impurities in isotopically enriched 28Si cooled below 10K, and compare two samples of different spin concentrations as a means of tuning the spin-spin interaction strength. The observations are consistent with a stable subharmonic peak at half the drive frequency which remains pinned even in the presence of pulse error for the strongly interacting sample. We propose a theoretical model based on the paradigmatic central spin model and show that the observations are consistent even in the presence of dissipation. Furthermore, a simple non-interacting Floquet dissipative model of a spin coupled to a bath appears remarkably close to the experiments. This suggests that the DTC order realised in solid-state experiments which are not fully isolated from the environment provide a novel realization of dissipative, semiclassical DTC order, and poses questions as to how to distinguish this phenomenon from the purely quantum effect.

Introduction: Symmetries provide an elegant way of classifying the various phases of matter realized in the physical world. Breaking of a symmetry is marked by the formation of order which gives rise to long range correlations in the system. The mechanism of spontaneous symmetry-breaking describes the phase transitions into equilibrium macroscopic states with long-range order. A commonly occurring broken-symmetry phase is a crystal in real space where the symmetry under continuous translation is broken to a lower discrete one. Until recently it was believed that an analogous phase which breaks time-translation symmetry and ‘crystallizes’ in the time direction, thereby forming a time crystal, is impossible. At thermal equilibrium, a macroscopic state without the breaking of spatial symmetry due to disorder fails to support these long-lived oscillation with a robust and well-defined frequency.

Many-body localization in strongly disordered, interacting, isolated quantum systems exhibiting breakdown of thermalization provides a novel mechanism for circumventing this fundamental limitation. Periodically driven many-body localized systems enable a route for breaking the discrete time-translational symmetry to a lower symmetry, where the macroscopic state oscillates with a longer time period, thereby forming a discrete time crystal. A generic mechanism for the breakdown of thermalization in systems with strictly local interactions is many-body localization (MBL). Although MBL is argued to be a necessary condition for the existence of time-crystallinity, recent observations of a discrete time crystal have been reported in systems with dipolar interaction and in the presence of a bath, which may not be many-body localized. These experiments can be interpreted to be in the prethermal or critical time-crystalline regime of isolated systems. There are even proposals to realize DTC order in cold atomic and quantum optical systems which are far from this limit.

In this article we report the observation of discrete time crystal formation in silicon doped with phosphorus (P). This experimental system had served as a motivation for many-body localization in Anderson’s seminal paper in 1958. High degree of quantum control of both the electronic spin states of P and the nuclear spin states of Si provide a unique platform for implementing dynamic pulse sequences crucial for realizing time crystals. On periodically driving the dopant spins, a stable DTC phase is observed in these experiments. In this article we present an alternate mechanism for stabilizing DTC behaviour in these systems. We go on to explore the periodically driven central spin model as a description for the observed DTC. This model is not many-body localized as the interactions are infinite range. We introduce dissipation to study the signatures of the DTC phase and show that this behaviour is consistent with experiments. Furthermore, we investigate the dynamics of a Floquet, dissipative non-interacting model which exhibits a striking similarity to the experimental data as well. Therefore, a semiclassical description of DTC in the presence of dissipation shares several qualitative features with the DTC order protected by many-body localization observed in earlier experiments.

Pulse protocols in doped silicon: A periodic pulse sequence is implemented on the electronic spins as shown in Fig. 1. We use a sample of 28Si enriched to 99.995%; this provides a magnetically clean environment which gives the dopant spins an exceptionally narrow linewidth of less than 10µT. The sample is placed in a Bruker X-band...
Spinlocking pulse $\pi$ $\pi/2$ $\pi$ $\pi$ 
Phase

a

+90°
0°

Hahn echo

Acquisition sequence

Floquet unitary (Repeat $N$ times)

b

FIG. 1. Microwave pulse control sequence and observation of time crystal phase. a: We initialise the spin ensemble with a $\pi/2$-pulse, then begin driving with $N$ Floquet unitaries, before applying a final $\pi/2$-pulse and read out with a $\pi/2 - \pi$ hahn echo sequence. b: We drive the spin ensemble with a fixed error of $\delta = -12$ ns and compare different values of interaction time $\tau_{int}$. For sufficiently long $\tau_{int}$, we see a single oscillation frequency that is stable to error. This is characteristic of discrete time crystalline order. Inset: comparison between two samples with and without DTC. The strength of the $\nu = 1/2$ oscillations decrease rapidly from their maximum as we change the error in the $\pi$ rotation pulse on sample A. In contrast, with sample B we see a clear plateau region where the $\nu = 1/2$ oscillations are stable to errors, indicating a DTC phase.

Experimental observations: We sweep the the number of Floquet cycles $N$ from 0 to 23 with error $\delta$ and map out the integrated echo amplitude as a function of $N$. The result is a decaying oscillating signal with frequency $\nu = 1/2$. We take the Fourier transform of these oscillations and examine what happens to the $\nu = 1/2$ peak as we change $\delta$. In the absence of interactions between electron spins, when we increase $\delta$ on the $\pi$-pulse in each Floquet cycle we expect the cumulative error caused by successive over- or under-rotation of the spin ensemble to cause modulation of the oscillations. This is apparent in the Fourier transform as a splitting of the $\nu = 1/2$ peak, as shown in Fig. 1b. Increasing $\delta$, we expect to see these peaks split further apart as the error in the $\pi$ rotations accumulates faster leading to faster modulation. However, in the presence of sufficiently strong interactions between neighbouring electron spins, we do not see an immediate splitting of the $\nu = 1/2$ peak, but instead find a region where the oscillations remain resilient to error.

In our experiment we tune the interactions in two ways. First, by changing the interaction time $\tau_{int}$, as shown in Fig. 1. Here we see the characteristic peak splitting due to a fixed 12 ns error in the $\pi$-rotations when the spins are allowed to interact for $\tau_{int} = 10\pi$. However, increasing the interaction time to $\tau_{int} = 30\pi$ we see the peaks move back together and at $\tau_{int} = 60\pi$ the $\nu = 1/2$ peak has fully returned, indicating that the oscillations are becoming more resilient to errors as we increase the interaction time.

In the inset to Fig. 1 we compare two samples with different concentrations of P dopant spins. Samples A and B contained concentrations of $1 \times 10^{15}$ cm$^{-3}$ and $3 \times 10^{15}$ cm$^{-3}$ respectively. $T_2$ was measured as 0.9 ms for sample A and 0.339 ms for sample B. This difference is due to the different spin-spin interaction strengths of the samples. We calculate the ‘crystalline fraction’ $\nu T_2$ as the ratio of the $\nu = 1/2$ peak to the total spectral power, $f = |S(\nu = 1/2)|^2 / \sum_{\nu} |S(\nu)|^2$ and plot this as a function of $\delta$ for the two samples shown in the inset to Fig. 1. We see that sample A has a clear peak at approximately $-6$ ns, either side of which the crystalline fraction falls off rapidly. In contrast, when we look at sample B we see a wide region between approximately $-12$ ns and $6$ ns error where the crystalline fraction remains approximately constant. This indicates the presence of a DTC phase with stability to error corresponding to $\pm 5\%$ of the perfect pi-pulse. We attribute the offset of the peak in crystalline fraction to an effect of the microwave pulse amplifier duty cycle.

We model the phenomenology of DTC behaviour observed in the experiments using the central spin model (CSM). The CSM has been successful as a semiclassical effective model for describing decoherence in solid state systems. The electron interacts with the nuclear spins of $^{28}$Si through the contact hyperfine interaction. Since the $^{29}$Si with $S = 1/2$ are randomly distributed, the coupling of the central spin to them is a source of random field on the electron spins. $^{28}$Si and $^{30}$Si, the other isotopes of Si occurring in the material are spinless, therefore significantly enhancing the coherence time of the electronic spins. We use the central spin model to de-
scribe the coherently driven dynamics of the ensemble of electronic spins. We expect this to provide an appropriate phenomenological description of the experimental system with long-range interactions and dissipation, the regime in which time crystalline behaviour has been observed.

\[
H_1 = \sum_i h_i \sigma_z^i + \sum_{j} J_{0,j} \sigma_z^i \sigma_z^j \tag{1}
\]

\[
H_2 = (\Gamma + \epsilon) \sum_i \sigma_z^i \tag{2}
\]

\( h_i \) is a random potential chosen from a uniform distribution between \([-\delta h, \delta h]\), while the width of the distribution of the interaction strength \( J_{0,j} \) is its mean value \( J \). \( N \) is the total number of spins in the system. In the experimental system considered in this article, the scale of the random potential \( h \) is \( \sim 224\text{kHz} \) while \( J \sim 165\text{Hz} \). In the rotating frame, the average field \( h \) is cancelled leaving only the random terms. For strong driving, in the rotating frame the flip-flop terms can be neglected with the interaction being diagonal in the \( \sigma_z \)-basis. The spinlocking pulse given by \( H_1 \) is applied for a time period \( \tau_{int} \) while the second pulse is applied for a period \( \tau_{flip} \)

\( \tau_{int} + \tau_{flip} = T = 1 \). We show that periodically driven CSM model has the essential ingredients to realise the physics of DTC. In the presence of dephasing which acts locally on the entire system, the DTC peak remains rigid.

The dephasing rate is given by \( \kappa = 1/T_2 \) which is the dominant mechanism in the relevant time scale of the experiment, acts uniformly on all the spins. By performing the Lindblad evolution of the density matrix, we study the dynamics of the central spin at the end of each time-period defined as:

\[
C_n = \langle \sigma_0^z(nT) \sigma_0^z \rangle \tag{3}
\]

The Fourier transform \( C_n \) is shown in Fig. 2b for a fixed rotation error \( \epsilon = 0.047^{-1} \) and two different interaction strengths. For strong interactions, a DTC peak at \( \nu = \nu_0/2 \) is robust to rotation errors, indicating the presence of DTC order, while for weaker interactions the peak splits. The subharmonic peak is indeed present for weaker interactions, yet the location of the subharmonic peak in frequency deviates with the pulse error while at stronger interaction strength the subharmonic peak remains pinned, a signature of DTC order (see Fig. 3). The peak at \( \nu = \frac{1}{2} \) is stable for a range of \( \epsilon \sim 0 - 0.047^{-1} \). This shows that interactions play a crucial role at stabilizing the DTC phase even in the dissipative regime. Interestingly, the lifetime of the DTC order also becomes longer in the strongly interacting regime, as shown in the inset to Fig. 3 further exemplifying the importance of interactions for this phenomena.

We also use the Bloch equations of a single spin coupled to a bath to model the dynamics of the spin magnetization. This is a rather simplified model but it exemplifies that even a non-interacting model bears resemblance to the experimental observations and highlights the pitfalls of

\[
\begin{align*}
\text{FIG. 2. a:} & \text{ A cartoon of the central spin model. b:} \text{ The behaviour of the spectral function for different interaction strengths } J \text{ at a fixed rotation error } \epsilon = 0.04. \text{ For strong interactions, the subharmonic peak at } \nu_{DTC} = \nu_0/2 \text{ is stable to finite rotation error } \epsilon. \text{ Both } \epsilon \text{ and } J \text{ are expressed in units of the inverse of the Floquet frequency. The dephasing time is chosen to be } T_2 = 50T. \text{ Inset: Comparison of the crystalline fraction as a function of rotation error for different interaction strengths. Increasing the interaction strength reduces the variation of the crystalline fraction with rotation error.}
\end{align*}
\]

\[
\begin{align*}
\text{FIG. 3. The position of the subharmonic peak as a function of rotation error } \epsilon \text{ for different interaction strengths. At weak interaction, the peak location drifts with pulse error while at stronger interaction the peak remains pinned, a signature of the DTC phase. Inset: The behaviour of the DTC peak width is shown as a function of rotation error } \epsilon. \text{ The different curves are for different interaction strength given in the legend. The DTC lifetime shows a parabolic dependence on pulse error close to } \epsilon \approx 0.
\end{align*}
\]
distinguishing many-body localized DTC order from this dissipative effect. The equations of motion of the single spin are

\[
\frac{d}{dt} \begin{pmatrix} M_x \\
M_y \\
M_z
\end{pmatrix} = \begin{pmatrix}
\frac{1}{T_1} & -\gamma B_y & -\gamma B_z \\
-\gamma B_z & \frac{1}{T_2} & \gamma B_x \\
-\gamma B_x & \gamma B_y & \frac{1}{T_1}
\end{pmatrix} \begin{pmatrix} M_x \\
M_y \\
M_z
\end{pmatrix} + \begin{pmatrix} 0 \\
0 \\
\frac{\mu_0 M_0}{\gamma B}
\end{pmatrix}
\]

where \( \mathbf{M} \) is the total magnetization, \( \mathbf{B} \) is the external magnetic field and \( \gamma \) is the gyromagnetic ratio. We assume we have an ensemble of non-interacting spins, modelled by a group of effective spins each governed by Eq. 4 with non-uniform response to the \( \theta \) rotation pulses due to an inhomogeneous \( B_1 \) field. This is captured by applying a Gaussian distribution of rotation pulses to the spins, centred around the target rotation angle \( \theta \) with a standard deviation \( \sigma \). The Gaussian is clipped such that no spins have more than 10% error. This models the finite extent of the sample. Thus after each \( \theta \) rotation the model spins are spread out in the Bloch sphere by some amount dictated by \( \sigma \) and the maximum allowed error. During the spinlocking pulse the spins precess around the spinlocking field vector. The damping terms in the Bloch equations, given by \( T_1 \) and \( T_2 \) (these are defined for the rotating frame), cause the spins to converge on the axis of the spinlocking vector, thus bringing them back into phase and correcting the error in the rotation pulses. We can see in Fig. 4 that this results in remarkably similar dynamics to the experimental data.

We can, however, identify some quantitative differences between the theoretical analysis and the experimental parameter regimes where the DTC order is realized. While \( T_1 \) was set to 130\( \mu \)s, which was measured as a lower bound for \( T_1 \), \( T_2 \) for the simulation was 20\( \mu \)s — based on variation in the microwave cavity field strength we would expect a value of approximately 1\( \mu \)s (however this value was not measurable in the experiment). The reason for the large difference in \( \sigma \) between the two samples requires additional investigation. Differences in sample geometry is a potential cause, however the size and shape of both samples were very similar. The simulation is unable to account for the behaviour of the crystalline fraction as a function of rotation error that we see in Fig. 1 if we use a smaller difference in \( \sigma \).

**Conclusions:** We have shown robust experimental signatures of the formation of discrete time-crystal phase in naturally purified silicon doped with phosphorus atoms. The experiments were performed on two samples with different dopant concentrations. We observe the formation of discrete time-crystalline order by driving the electron spin ensemble at frequency \( \nu_0 \), while producing a response at a sub-harmonic frequency \( \nu_0/2 \). This peak remains pinned and is robust to perturbations in the periodic pulse protocol. Motivated by the experimental system, we investigate the dissipative central spin model as a phenomenological description for time-crystalline behaviour in solid state systems with long-range interactions. We show that the model demonstrates the essential features measured in the experiments, a DTC peak which is stable at finite pulse error appears for strong enough interactions. We further examine the extent to which these phenomena can be modelled by non-interacting spins coupled to a bath and find that several key features of the data can be recreated in this model. The difference in standard deviation in pulse error between samples for which this effect is realized in the model is significantly different from what we would expect in the experimental system.

The high level of quantum control of electron and nuclear spins in phosphorus doped silicon provides a promising platform for studying many-body quantum coherence in driven systems. By driving the nuclear and electronic spins independently, proximity effects in time-crystalline behaviour and effects of dynamic nuclear polarization can be explored. The difference in the dynamic time scales of electron and nuclear spins could serve as a useful tool for manipulating DTC order and its long time coherence.
Furthermore, the life time of the time crystal order can be exploited to probe the dephasing and thermalizing properties of long range systems. The DTC order exhibited in the central spin model poses several interesting questions for further investigation. The distinction between many-body localized and dissipative, semiclassical DTC order is an important question, which the CSM model may be able to shed some light on. However, given the striking similarity between the experimental data and our semiclassical analysis, it is salient to find observables which capture the quantum to classical crossover. Experimental measurements of quantum correlations in space and time can provide a precise distinction between the two regimes.

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