Freezing a Quantum Magnet by Repeated Quantum Interference: An Experimental Realization

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We experimentally demonstrate the phenomenon of dynamical many-body freezing in a periodically driven quantum Ising magnet within an NMR simulation scheme. The phenomenon is essentially a result of repeated quantum interference between the amplitudes of the fundamental excitations of the many-body system. The degree of freezing exhibits surprising non-monotonic behavior with respect to the driving frequency. At the points of maximal freezing, the population dynamics of all the quasi-particle modes are frozen very strongly for all time, which renders the freezing occurring independent of initial state and visible for almost all system sizes. Magnetization measured for our finite spin system gives direct access to the time-evolution of the underlying fermionic excitations in momentum space.

The study of non-equilibrium dynamics has emerged as one of the central topics in quantum many-body physics, thanks to the recent advancements in various experimental techniques in isolating and coherently controlling quantum systems over fantastically long time scales [1–3]. In particular, the paradigm of periodically driven many-body systems has hosted quite a few interesting theoretical results recently (see, for example, [4–9]). Coherent periodic driving has also gathered significant experimental attention (see, e.g., [10–13]) in recent days, bringing up the topic as one of the focal interests of quantum nonequilibrium physics. For example, the average Hamiltonian theory developed and practiced by Waugh and co-workers, founded high-resolution solid state Nuclear Magnetic Resonance (NMR) [14]. It involves systematic coherent driving of spin systems, via a time-dependent Hamiltonian, leading to an effective evolution under desired interactions. A constant drive with a strong transverse field, called spin-lock, is used in many of the NMR experiments to create an effective transverse Hamiltonian in the rotating frame [15]. We add to this list, experimental verification of a surprising non-equilibrium freezing phenomenon where quantum interference leads to almost absolute freezing of a simple magnet under periodic drive for certain discrete values of the of the driving frequency and the amplitude [16].

In general, response of a simple driven system may “freeze” if the drive rate is much faster compared to the characteristic relaxation rate of the system. The mechanism is intuitively simple – the system does not get sufficient time to change itself significantly within the duration of the drive. Thus, faster the drive is, more frozen is the response. This intuition is manifested in many celebrated results in both classical and quantum physics, like Kibble-Zurek scaling law for defect generation in classical [17, 18] and quantum [19–21] phase transitions, Landau-Zener excitation probability [22, 23], phenomenon of classical dynamical hysteresis [24] to name a few. It forms the basis of the so called sudden approximation in quantum mechanics [25], and its more elaborate descendant adiabatic-impulse approximation which applies to classical and quantum systems alike [18, 26]. For a periodically driven system, this intuition implies a stronger freezing for a higher frequency of the drive. However, in presence of quantum interference this powerful intuitive picture may grossly fail to predict the dynamics, as the dynamics depends not on the probabilities of transitions but on their amplitudes – suppression (enhancement) of the transitions due to destructive (constructive) quantum interference may play a major role [27]. In our experiment we demonstrate one such phenomenon, namely, dynamical many-body freezing, where repeated quantum interference results in strongly non-monotonic freezing behavior with respect to the driving rate in a periodically driven quantum magnet, contrasting the above intuitive scenario [16, 28–32]. Most surprisingly, absolute freezing (freezing of all quasi-particle excitations) is observed for certain particular values of drive parameters in a large class of integrable models for various kinds of periodic drive [16, 28, 29]. Quantum behavior being pivotal in the freezing phenomenon (see [16, 27]), its experimental demonstration poses a major challenge, as decoherence limits the experimental time-scale (particularly in presence of the external drive). We diminish this hurdle in our present work by choosing a system with a long coherence time. For a many-body system, decoherence is more severe as the system-size is increased. But in our case, since all degrees of freedoms (not only the low energy modes) freeze at certain discrete frequencies (freezing peaks), the location of the peaks and the qualitative features predicted by the theory for infinite system, can be captured quite accurately even with systems of mini-
The rest of the paper is organized as follows. First, we outline the theory and the phenomenology of the dynamical many-body freezing, particularly for our finite-size experimental system, then we discuss the experimental setup and realization of the phenomenon followed by the result and its analysis, and finally we conclude with an outlook.

**Freezing in a 3-spin Ising Chain** – We choose to study a one-dimensional Ising ring that is being subjected to a transverse periodic-driving field of amplitude $h_0$, frequency $\omega$, and represented by the Hamiltonian [16],

$$\mathcal{H}(t) = -\frac{1}{2} \left( J \sum_{i,j=1}^{3} \sigma_i^x \sigma_j^x + h_0 \cos(\omega t) \sum_{i=1}^{3} \sigma_i^z \right). \quad (1)$$

Here $J$ is the strength of the nearest neighbor Ising interaction and $\sigma_i^{x/z}$ are Pauli matrices. We use dynamical order parameter $Q$ [16], defined as the long-time average of magnetization in $x$-direction, $m^x(t)$, to characterize freezing via the anomalous DC response,

$$Q = \lim_{T\to\infty} \frac{1}{T} \int_0^T m^x(t) dt, \quad (2)$$

where $T$ is the duration of total evolution. Suppose that the system begins from the completely polarized state in the $x$-direction. Then the adiabatic limit ($\omega/L \to 0$), where $L$ is the system-size) corresponds to $Q = 0$ in $L \to \infty$ limit (since the average of the transverse field is zero over each cycle), and the impulse or “freezing” limit of $(\omega \to \infty)$ corresponds to $Q = 1/2$.

Classical intuition suggests a monotonic behavior of $Q$ as a function of $\omega$ interpolating between these two regimes. But quantum interference leads to a strongly non-monotonic freezing behavior which, for large $\omega$, can be accurately given by a closed form analytical formula for $Q$:

$$Q = \frac{1}{1 + |J_0(2h_0/\omega)|} \quad (3)$$

in $L \to \infty$ limit [16], where $J_0(.)$ denotes the ordinary Bessel function of order 0. The above result derives (for full $+x$ initial polarization) as follows [16, 27]. The Hamiltonian (11) can be mapped to independent fermionic Hamiltonians in momentum space such that only fermions with equal but opposite momenta $\pm k$ are coupled (see e.g.[31]). The Hilbert space $\mathcal{H}$ for the chain is consequently factorized into a direct product of $L/2$ two-dimensional Hilbert spaces $\mathcal{H}_k$ for each momentum $k > 0$ and the wave function of the chain takes the form $|\psi(t)\rangle = \otimes_{k>0} (u_k|0_k,0_{-k}\rangle + v_k|1_k,1_{-k}\rangle)$, where the basis states $\{|0_k,0_{-k}\rangle, |1_k,1_{-k}\rangle\}$ correspond respectively to the completely vacant and fully occupied $\pm k$ states.

The dynamics of $m^x$ translates to the population dynamics of the independent two level fermionic systems: $m^x(t) = \frac{1}{2} \sum_{k>0} |v_k(t)|^2 - 1; |v_k|^2 = 1 - A_k \sin^2(\phi_k t)$, where the response amplitude is $A_k = (\Delta_k/\phi_k)^2$, with $\Delta_k = J \sin(k) J_0(2h_0/\omega)$, and the response frequency is

$$2\phi_k = 2 \sqrt{\Delta_k^2 + (J \cos k)^2}. \quad (4)$$

Absolute freezing (under rotating wave approximation) happens when $A_k$ vanishes for all $k$ under the condition

$$J_0(2h_0/\omega) = 0. \quad (5)$$

For $L = 3$, the $k$ values are $\{-\pi, \pm \pi/3\}$, leading to

$$m^x_{L=3}(t) = 1 - 4/3(\Delta_k/\phi_k)^2 \sin^2(\phi_k t); \quad k = \pi/3. \quad (6)$$

We simulate the above freezing phenomenon in the 3-spin chain given by Eq. (11) using the three spin-$1/2$ $^{19}$F nuclei of trifluoriodoethylene dissolved in acetone-D6. The molecular structure of the system is shown in Fig. 1(a). All the experiments described below were carried out using a Bruker 500 MHz NMR spectrometer at an ambient temperature of 290 K. The simulation involves preparing a certain initial state, evolving it under the Hamiltonian $\mathcal{H}(t)$ for a total time $T$, while measuring the response of the system in intervals of time $\tau$.

The thermal equilibrium state for the NMR spin-system is $\sum_{i=1}^{3} \sigma_i^z$ [27]. The response of the system measured as transverse magnetization is given by

$$m^x(t) = \frac{1}{m_0} \text{Tr} \left[ \rho(t) \left( \sum_{i=1}^{3} \sigma_i^x / 2 \right) \right]. \quad (7)$$

Here $\rho(t)$ is the instantaneous density matrix of the driven system and $m_0$ is a normalization factor [27] given by the maximum possible transverse magnetization i.e. $m_0 = \text{Tr} \left( \sum_{i=1}^{3} \sigma_i^x \sum_{i=1}^{3} \sigma_i^x / 2 \right) = 12$. We performed a set of experiments for each of the following two different initial states. The first initial state is $\rho(0) = \sum_{i=1}^{3} \sigma_i^x$ (i.e. $m^x(0) = 1$, fully polarized in $+x$-direction) obtained by applying a global $R_y(\pi/2) = \exp(-i\pi/2) \sum_i \sigma_i^y / 2$
rotation on the thermal state. This state is approximately the ground state of our Hamiltonian (11) at \( t = 0 \), if \( h_0 \gg J \). The second initial state is \( \rho(0) = \sum_{i=1}^{3} (\sigma_i^x / 2 + \sigma_i^z \sqrt{3}/2) \) (i.e. \( m^x(0) = 0.5 \)) obtained by applying a global \( R_y(\pi/6) \) rotation on the thermal state. Note that this state is not the ground state of Hamiltonian (11). For convenience, we refer these two sets of experiments by their initial magnetization values, \( m^x(0) \).

The internal Hamiltonian for the NMR system consists of the Zeeman part and the spin-spin interaction part:

\[
\mathcal{H}_{\text{int}} = -\pi \sum_{i=1}^{3} \nu_i \sigma_i^z + \frac{\pi}{2} \sum_{i,j=1}^{3} J_{ij} \sigma_i^z \sigma_j^z. \tag{8}
\]

Here \( \nu_i \) is the resonance frequency of the \( i \)th spin in the rotating frame, and \( J_{ij} \) is the strength of the indirect spin-spin interaction between spins \( i \) and \( j \). The parameters of internal Hamiltonian for the above spin-system are shown in Fig. 1 (b).

In order to simulate the time-dependent Hamiltonian \( \mathcal{H}(t) \) using \( \mathcal{H}_{\text{int}} \), we need to (i) cancel the evolution under Zeeman interaction, (ii) bring out an effective Ising interaction of strength \( J \), and (iii) add the oscillatory drive \((-h_0/2) \cos(\omega t)\) along the x-direction. We discretize the time into \( M \) equal steps each of duration \( \delta t = T/M \). The propagators for the discretized Hamiltonian are

\[
U_k = \exp[-i\delta t \mathcal{H}(k\delta t)], \tag{9}
\]

for \( k \in \{1, 2, \cdots, M\} \). The overall evolution for a time \( k\delta t \) is given by the unitary \( U(k\delta t) = U_k U_{k-1} \cdots U_2 U_1 \).

The response \( m^x(t) \) (refer Eq. (7)), is measured at time instants \( t = n\tau \), for \( n = 0, 1, 2, \cdots, N \), where \( N = T/\tau \) corresponds to the last measurement. Therefore we only need to generate the propagators \( U(n\tau) \). We used Gradient Ascent Pulse Engineering (GRAPE) method to design amplitude and phase modulated radio frequency (RF) pulses which effectively realize these propagators [33].

The dynamical order parameter for this discretized set of measurements is defined as

\[
Q = \frac{1}{N+1} \sum_{n=0}^{N} m^x(n\tau). \tag{10}
\]

The experiments were carried out with 14 distinct \( \omega \) values ranging from 3.59 to 24.54 rad/s. In each case we chose the measurement interval as \( \tau = 2\pi/\omega \), the period of the driving frequency. We discretized each cycle of the periodic Hamiltonian \( H(t) \) into 11 steps and constructed the propagators \( U(n\tau) \) for \( N = 30 \) cycles, i.e., \( n = 0, 1, 2, \cdots, N \). Here maximum number of cycles was limited by the duty-cycle of the RF channel.

The one-cycle propagator \( U(\tau) = U_{11} U_{10} \cdots U_2 U_1 \), where \( U_{ks} \)s are given by Eq. 9. Since \( H(t) \) has a period of \( \tau \), \( U(n\tau) = U^n(\tau) \). All numerically generated GRAPE pulses were optimized against RF inhomogeneity and had an average Hilbert-Schmidt fidelity greater than or equal to 0.99. For the 30th cycle, the overall duration of the GRAPE pulses were ranging between 48 ms to 300 ms for different \( \omega \) values.

The overall experimental sequence can be represented by the unitary \( U(n\tau) \cdot R_y(\pi/2) \) for \( m^x(0) = 1 \) and \( U(n\tau) \cdot R_y(\pi/6) \) for \( m^x(0) = 0.5 \). The net-transverse magnetization corresponding to the observable \( \sum_{i=1}^{3} \sigma_i^x / 2 \) was measured as the intensity of the real part of the NMR signal acquired in a quadrature mode and is normalized by the reference signal obtained with only \( R_y(\pi/2) \) pulse [27].

The main result of our experiment is summarized in Figs. 2, 3 and 4. In Fig. 2, dynamics of \( m^x \) as the function of time is shown for a few values of \( \omega \), and for parameters \( h_0 = 5\pi \) and \( J = h_0/20 \) (in Hz). The freezing of the dynamics is clearly visible for the theoretically predicted values of \( \omega \). The oscillation of \( m^x(t) \) allows us to directly read off the frequency of the population oscillation of the underlying fermionic mode \( k = \pi/3 \) in the momentum space using Eq. (6). However, from the raw experimental data (green line-filled-circle), we see \( m^x \) decays steadily even under the maximal freezing condition.

![FIG. 2.](https://example.com/figure2.png)  
(Color online) Dynamics of \( m^x \) vs \( t/\tau \) (\( \tau = 2\pi/\omega \)) for \( m^x(0) = 1 \). Strong freezing behavior is observed for theoretically predicted values: \( \omega = 5.61 \) and 12.88 rad/s, while substantial oscillations are visible even at larger driving frequencies \( \omega = 8.40 \) and 24.54 rad/s. One can directly read off the frequency of population-oscillation of the Jordan-Wigner fermion (for \( k = \pi/3 \)) from the plot. The period of oscillation of \( m^x \) visible from the experimental data (marked for \( \omega = 8.40, 24.54 \) rad/s) corresponds to the frequency \( 2\nu_0 \) of population oscillation of the underlying fermionic quasiparticle with momentum \( k = \pi/3 \) given in Eq. (4). The analytical curves represent Eq. (6) which establishes this correspondence.
of an infinite chain driven over infinite time!

Interestingly, not only the freezing peaks, but the theoretically predicted freezing conditions given by the analytical formula in main same. We also compare the finite size result in Fig. 3 with inverse decay. Experimental results (e), (f) Experimental results.

\( \omega = 3.59, 5.61, \text{and } 12.88 \text{ rad/s} \). This decay is due to the transverse relaxation \((T_{1\rho}) [34]\) as well as incoherence caused by the spatial inhomogeneities in RF amplitudes [15]. For all other values of \( \omega \), within our chosen \( \omega \) range, we see oscillations in \( x \)-magnetization in addition to the decay. In order to confirm this, we took into account the effect of decay by multiplying the experimental data points \( m^x(t) \) with the standard inverse decay functions, i.e., \( \exp(t/T_2) \). Here the decay constant \( T_2 \) is obtained by the exponential fitting of \( m^x(t) \). The resulting data points are shown in Fig. 2 as cross connected with solid lines.

Fig. 3 shows \( Q \)-vs-\( \omega \) plots again for experimental parameters \( h_0 = 5\pi \) and \( J = h_0/20 \) (in Hz). The peaks represent the freezing points. We observe three freezing points in this range, viz. at 3.59, 5.61, and 12.88 rad/s (as indicated by dashed vertical lines). Comparison of the experimental data after inverse-decay correction (Fig. 3 (e),(f)) with the exact numerical simulation for the \( Q \)-lines exhibits striking agreement. The raw experimental data in Fig. 3 (g),(h) exhibits an over-all downward shift of the experimental line due to the \( T_2 \) decay, though the basic features (particularly, the peak positions) remain same. We also compare the finite size result in Fig. 3(c),(d) with the infinite size analytical formula (3) in Fig. 3(a),(b). The freezing points match accurately with the theoretically predicted freezing conditions given by Eq. (5).

Interestingly, not only the freezing peaks, but also the trend of most of the \( Q \) vs \( \omega \) profile for our 3-spin system driven over 30 cycles matches fairly well with that of an infinite chain driven over infinite time!

In Fig. 4, we show the experimentally obtained spectra for \( m^x(0) = 1 \) at time instants \( 0, 3\tau, 6\tau, \cdots, 30\tau \), for two driving frequencies (i) \( \omega = 24.54 \) rad/s not satisfying the freezing condition (left column) and (ii) \( \omega = 5.61 \) rad/s satisfying the freezing condition (right column). The spectra at time instant \( n = 0 \) correspond to the state completely polarized in \( +x \) direction. The phase-oscillations of the the spectral lines in the non-freezing case (left column) can be clearly noticed, while the freezing spectra (right column) remain in-phase.

**Conclusion and Outlook** — In our NMR experiments we have demonstrated that repeated quantum interference can strongly freeze the magnetization dynamics in of a periodically driven Ising chain for any initial state for certain particular values of the driving amplitude and the frequency, and confirm the phenomenon of dynamical many-body freezing. The novelty of the phenomenon arises from its departure from the standard picture of periodic drive close to the resonance or in the perturbative limit, since it lives in a highly off-resonant regime \( (\omega \gg 2J) \) and under strong drive \( (h_0 \gg J) \). In the field of NMR, freezing spin dynamics (spin-locking) using a strong constant drive along the spin-polarization axis is a well known and routinely used technique for suppressing unwanted evolutions. Cosine modulated spin-lock was earlier used in NMR for several applications such as excitation of multiple transitions in quadrupolar spin systems [35] and selective coherence transfers in homonuclear dipolar coupled spin systems [36]. The major contribution of the present work in this respect is to extend such control over quantum state evolution to
many-body level, which allowed us freezing the many-body response for arbitrary initial state by freezing all the collective excitations. Further investigations of the freezing phenomenon in larger spin systems and under different types of interactions will be interesting. With further experimental accuracies, complete freezing of certain quantities against unwanted evolution in a system of interacting qubits (Ising spins) might be possible by imposing strong Ising interactions between the qubits and a suitable periodic drive.

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SUPPLEMENTARY MATERIAL

DYNAMICS OF THE PERIODICALLY DRIVEN ISING RING FOR \( L = 3 \)

The Hamiltonian we consider is

\[
\mathcal{H} = -\frac{1}{2} \left[ J \sum_{i} \sigma_{i}^{x} \sigma_{i+1}^{x} + h_{0} \cos(\omega t) \sum_{i} \sigma_{i}^{z} \right],
\]

(11)

with \( L = 3 \) in our case and Periodic Boundary Condition \( \sigma_{4} = \sigma_{1} \). The eigen-problem of the above Hamiltonian can be solved analytically by Jordan-Wigner transformation followed by Fourier transform for any \( L \) (see e.g., [37, 38]). The Hamiltonian (11) can be mapped to independent fermionic Hamiltonians in momentum space such that only fermions with equal but opposite momenta \( \pm k \) are coupled:

\[
\mathcal{H}(t) = \bigotimes_{k>0} H_{k}; \quad H_{k} = -E_{k}(c_{k}^{\dagger}c_{k} - c_{-k}^{\dagger}c_{-k}) + i\Delta_{k}(c_{-k}c_{k} - c_{k}^{\dagger}c_{-k}),
\]

(12)

where \( E_{k} = h_{0} \cos \omega t + J \cos k \), \( \Delta_{k} = J \sin k \), with \( \delta = J_{x} + J_{y} \) and \( \gamma = J_{x} - J_{y} \), \( h_{0}(t) = h_{0} \cos \omega t \). The quantization of \( k \) depends on the parity of the fermion number (i.e., whether it is odd/even) which is conserved throughout the dynamics. Here for definiteness, we start with the ground-state of \( \mathcal{H} \) for any parameter value, which will always have even fermionic number. This leads to the quantization (modulo the choice of the Brillouin zone) \( k = -\pi, \pm \frac{(2n+1)\pi}{L} \), \( n = 0, 1, 2, ..., \frac{L-3}{2} \). In our case, we chose have \( \{-\pi, -\pi/3, \pi/3\} \). Now we note that \( k = -\pi \) has no partner to pair with, while the only pair formed is for \( k = \pm \pi/3 \). Thus we can work with a Hilbert space spanned by the vectors \( \{|0_{\pi/3},0_{\pi/3}\}, |1_{\pi/3},0_{\pi/3}\rangle \otimes |0_{\pi/3},1_{\pi/3}\rangle \). In this representation the initial state fully polarized in \( +x \)-direction reads \( |\psi(0)\rangle = |1_{\pi/3},1_{\pi/3}\rangle \otimes |1_{\pi/3}\rangle \). Henceforth we denote \( |0_{\pi/3},0_{\pi/3}\rangle \) by \( |0_{\pi/3}\rangle \) and \( |1_{\pi/3},1_{\pi/3}\rangle \) by \( |1_{\pi/3}\rangle \). Noting that the \( k = -\pi \) mode has no dynamics, the state at any time can be given by

\[
|\psi(t)\rangle = [u_{\pi/3}|1_{\pi/3}\rangle + v_{\pi/3}|0_{\pi/3}\rangle] \otimes |1_{\pi/3}\rangle,
\]

(13)

The time-dependent transverse magnetization \( m^{x}(t) = \frac{1}{2} \sum_{i} \sigma_{i}^{x} = \frac{1}{2} \sum_{k} (2c_{k}^{\dagger}c_{k} - 1) \) in this notation reads (for the initial state fully polarized in \( +x \)-direction),

\[
m^{x}(t) = \frac{4}{3} |v_{\pi/3}(t)|^{2} - \frac{1}{3}
\]

(14)

Now \( \{u_{\pi/3}, v_{\pi/3}\} \) satisfies following time-dependent Schrödinger equation [39]:

\[
\frac{i}{\hbar} \frac{\partial}{\partial t} \begin{bmatrix} u_{\pi/3}(t) \\ v_{\pi/3}(t) \end{bmatrix} = \begin{bmatrix} h_{0} \cos(\omega t) + J \cos \pi/3 & iJ \sin \pi/3 \\ -iJ \sin \pi/3 & -h_{0} \cos(\omega t) - J \cos \pi/3 \end{bmatrix} \begin{bmatrix} u_{\pi/3}(t) \\ v_{\pi/3}(t) \end{bmatrix}
\]

(15)

Exact solution of the above \( 2 \times 2 \) matrix equation is not known but analytical solution can be obtained under a rotating wave approximation in the fast driving regime \( (\omega \gg 2J) \) [16]. For the initial condition mentioned above, the solution for \( |v_{\pi/3}|^{2} \) reads

\[
|v_{\pi/3}|^{2} = 1 - A_{\pi/3}^{2} \sin^{2}(\phi_{\pi/3}t),
\]

(16)

where \( \phi_{\pi/3} = \sqrt{\Delta_{k}^{2} + J^{2} \cos^{2} \pi/3} \), and \( A_{\pi/3} = (\Delta_{\pi/3}/\phi_{\pi/3})^{1/2} \). The time-dependent magnetization and \( Q \) thus reads

\[
m^{x}(t) = 1 - \frac{4}{3} A_{\pi/3}^{2} \sin^{2}(\phi_{\pi/3}t); \quad Q = 1 - \frac{2}{3} A_{\pi/3}^{2},
\]

(17)

which are not monotonic functions of \( \omega \) and \( h_{0} \).
FIG. 5. Plot of energies of the instantaneous ground state (denoted by $|G\rangle$) and excited state ($|Ex\rangle$) of the $2 \times 2$ Hamiltonian (Eq. 15) as a function of $\omega t$. Our first sweep ($m = 0$) starts from the left end with the initial state $|\psi(0)\rangle = \{u_{\pi/3}(0) = 0; v_{\pi/3}(0) = 1\} \approx |G(0)\rangle$. $P_{ex}$ denotes the probability that starting from this initial state the system ends up with the same state (i.e. $\{u_{\pi/3} = 0; v_{\pi/3} = 1\}$) at the end of half a cycle, instead of following the drive adiabatically and ending up with the ground state ($\approx |0_{\pi/3}\rangle$) of the final Hamiltonian.

FIG. 6. Comparison of the results obtained by repeated calculation of transition probabilities instead of transition amplitudes (black line-triangle) with the exact numerical result (red line-diamond) and the experiment (blue line-star). Neglecting the phases after each half-cycle leads to a qualitatively different result even within the finite number of sweeps: while the experimental results matches qualitatively well with the theoretically predicted peak-valley structure of the $Q$ vs $\omega$ curve, the probability based calculation gives a monotonic behavior for the same. Inset shows Landau-Zener type excitation probability $P_{ex}$ for half-a-sweep as a function of $\omega$ calculated numerically. The results correspond to $h_0 = 5\pi$, $J = h_0/20$ (in Hz).

**PROBABILITY VS AMPLITUDE**

The non-monotonic nature of the freezing phenomena, including the appearance of maximal freezing peaks are consequence of quantum interference, as indicated in [16]. Here we explicitly demonstrate the role of quantum interference behind the key features of the freezing behavior. To this end we analyze the problem using another approach based on repeated calculation of Landau-Zener like excitation, which has been employed successfully to estimate certain aspects of a repeated quench dynamics in a similar model in presence of decoherence after each sweep [40]. We demonstrate how such an approach (based on counting of probabilities rather than amplitudes) fails to explain even the qualitative features of the freezing phenomenon. Suppose we start from the initial state...
ground state (to a good approximation) corresponds to the half-cycle. The name excitation probability is motivated by the fact that since \( h kT \), the high-temperature and high-field approximation (see Fig. 5). Now we want to estimate the probability \( |v_{\pi/3}(2m+1)|^2 \) of being in the state \( |1_{\pi/3} \rangle \) after the subsequent (i.e. \( 2m + 1 \)-th) half-sweep is made. We can come up with an estimate based on calculating transition probabilities if we are supplied with the excitation probability \( P_{ex} \), which is the probability that we start with the states \( |1_{\pi/3} \rangle \) before starting a half-cycle and end up with the same state \( |1_{\pi/3} \rangle \) after completing the half-cycle. The name excitation probability is motivated by the fact that since \( h_0 \gg J \), for \( h^x = +h_0 \) the ground state (to a good approximation) corresponds to \( |1_{\pi/3} \rangle \) (left end of Fig. 5), while for \( h^x = -h_0 \) the ground state approximately corresponds to \( |0_{\pi/3} \rangle \) (right end of the Fig.). Due to the symmetry of the problem, this is also the probability that we end up in the ground state starting from the excited state. Since \( h_0 \gg J \) cos \( k \), we can approximately use the same \( P_{ex} \) for the reverse sweep (using a different one for the reverse sweep doesn’t change the conclusion in any qualitative way). Now taking the excitation probabilities into account we find the following recursion relation

\[
|v_{\pi/3}(2m+1)|^2 = |v_{\pi/3}(2m)|^2 (2P_{ex} - 1) + 1 - P_{ex},
\]

where \( m = 0 \) denotes the initial state (starting from the left). Solving above relation one gets

\[
|v_{\pi/3}(2m+1)|^2 = \frac{1}{2} + (2P_{ex} - 1)^{2m} |v_{\pi/3}(0)|^2 - \frac{1}{2}
\]

We thus see that \( |v_{\pi/3}(2m+1)|^2 \to \frac{1}{2} \) as \( m \to \infty \) irrespective of \( \omega \). This in turn implies \( Q \to \omega \)-independent constant as \( m \to \infty \). In our case (\( L = 3 \)), this value is 1/3 (in general \( Q \sim 1/L \)). Thus the very fact that \( m^x \) acquires an anomalous steady value dependent on \( \omega \) and \( h_0 \) is a result of repeated quantum interference between the phases gathered after the half cycles.

Even for a finite number of sweeps realized in our present experiment, we see a qualitative difference between the experimental results (which agrees with the exact quantum treatment) and the above probability based calculation. In Fig. 6 we show an explicit comparison of the experimental result with the numerical result predicted by Eq. (19), carried out for the experimental parameters for 30 cycles. We calculated \( P_{ex} \) numerically exactly by evolving the system for half-a-cycle for each value of \( \omega \). \( m^x(n) \) is measured after each complete cycle and averaged over to calculate \( Q \) (as is done in the experiment). The result clearly shows a monotonic \( Q \) (black line-triangles) as a function of \( \omega \), in accordance with the classical idea, but in stark contrast with the full quantum theory (red line-diamonds) and experimental results (blue line-stars). This monotonic variation of \( Q \) estimated over finite cycles is clearly the consequence of the fact that \( P_{ex} \) is a monotonic function of \( \omega \) (see inset of Fig. 6) in our case, as one may expect from the classical adiabatic-impulse scenario [18].

**THERMAL EQUILIBRIUM STATE IN NMR**

The NMR density matrix of an ensemble of a system of \( n \) spin-1/2 homonuclear Nucleus (i.e. having same gyromagnetic ratio \( \gamma \)) in thermal equilibrium at an ambient temperature \( T \) and inside a uniform magnetic field \( B_0 \) in \( z \)-direction is [15]

\[
\rho = \frac{1}{Z} \exp(-\mathcal{H}/kT).
\]

Here

\[
\mathcal{H} = h\gamma B_0 \sum_{j=1}^{n} I_j^z,
\]

is the Hamiltonian , \( Z = \text{Tr}[\exp(-\mathcal{H}/kT)] = 2^n \) is the partition function and \( k \) is the Boltzmann Constant. Under the high-temperature and high-field approximation \( (kT \gg \Delta E) = h\gamma B_0 \), the energy gap), the above form can be expanded to

\[
\rho \approx \frac{1}{2^n} 1 - \mathcal{H}/kT \\
= \frac{1}{2^n} 1 + \epsilon \rho_\Delta,
\]

(22)
where $\epsilon = -\gamma hB_0/kT$, and $\rho_\Delta = \sum_{j=1}^{n} I_j^z$ is the traceless deviation part.

Since the Identity part does not transform under any Unitary transformation and does not give any signal in NMR, we ignore it. So in all practical calculations, we simply take the trace-less part $\rho_\Delta = \sum_{j=1}^{n} I_j^z$ or $\rho_\Delta = \sum_{j=1}^{n} \sigma_j^z$ (absorbing 1/2 in $\epsilon$) as thermal state NMR density matrix.

**SIGNAL IN NMR**

The real part of the signal (i.e. the magnetization along the $x$ direction), $S(t)$ in NMR is [15]

$$S(t) \propto \text{Tr}[\rho_t D], \quad (23)$$

where $\rho_t = U_t \rho_\Delta U_t^\dagger$ is the instantaneous density matrix after the Unitary transformation given by $U_t$, and the detection operator $D = \sum_{j=1}^{n} I_j^z$. To get rid of the proportionality constant, we normalize the signal with respect to the signal obtained after applying a global $(\pi/2)_y$ pulse on the thermal equilibrium state (i.e. $\rho_t = \sum_{j=1}^{n} \sigma_j^z$).

$$S_{\text{norm}}(t) = \frac{\text{Tr}[\rho_t D]}{\text{Tr}[\sum_{j=1}^{n} \sigma_j^z D]} = \frac{1}{12} \text{Tr}[\rho_t D] \quad \text{for } n = 3. \quad (24)$$