Experimental Implementation of Efficient Quantum Pseudorandomness on a 12-spin System

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Quantum pseudorandomness, also known as unitary designs, comprise a powerful resource for quantum computation and quantum engineering. While it is known in theory that pseudorandom unitary operators can be constructed efficiently, realizing these objects in realistic physical systems can be a challenging task. In this work, we study quantum pseudorandomness generation on a 12-spin nuclear magnetic resonance system. The experimental process is based on the recently proposed design Hamiltonian approach, which has the merit of being significantly more efficient than previous protocols. By applying random refocusing sequences to the experimental system we create a design Hamiltonian the dynamics of which quickly forms unitary designs. We then use multiple-quantum techniques to measure spreading of quantum coherences over system’s degrees of freedom, and so to probe the growth of quantum pseudorandomness. The measured multiple-quantum coherence spectra indicate that substantial quantum pseudorandomness have been achieved.

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Quantum randomness plays a significant role in quantum information science. It is a fundamentally important resource in quantum tomography [1, 2], noise characterization [3, 4], quantum chaos [5–7], quantum metrology [8], and many other areas. However, similar to the classical case, the complexity of generating fully random transformations on a quantum system grows exponentially with the system size [9]. Therefore, quantum pseudorandomness, often cast as unitary designs more formally, was put forth as an alternate. Unitary designs are operationally useful sets of unitaries—a k-design is any ensemble of unitaries capable of simulating up to the k-th order statistical moments of the Haar ensemble on average [10]. Recently, great efforts have been devoted to identifying efficient constructions of k-designs and to exploring their practical uses. In particular, unitary 2-designs were intensely studied, and were found to have efficient constructions either exactly from the Clifford group [11] or approximately from random quantum circuits [12–17]. However, in experimental aspects, progress is quite limited as unitary designs have been achieved only in small-sized physical systems [18–21]. As the scale of controllable quantum systems continues to grow rapidly today, realizing pseudorandom operations on these systems becomes an important and challenging task.

In this Letter, we study experimental generation of approximate unitary designs on a 12-qubit spin system, using techniques of nuclear magnetic resonance (NMR). On the whole, our study has to address two important problems. The first problem concerns experimental feasibility. There have been devised a variety of generation protocols that use, e.g., polynomial-sized random quantum circuits [12–17], graph state techniques [22, 23], or random dynamics of design Hamiltonian [24]. With feasible experimental realization in mind, we follow the design Hamiltonian approach in our work due to its benefits such as saving of qubit resources and reducing of time cost, compared with the other protocols. A design Hamiltonian is some random Hamiltonian satisfying that its time-evolutions form unitary designs spontaneously. Actually, a concrete form of design Hamiltonian already appeared in Ref. [24], which is composed of periodically changing random spin-glass-type interactions. Here, we show that these disordered interactions can be readily simulated by means of NMR refocusing techniques. Our numerical and experimental results indicate that, evolving the 12-qubit system under a suitably created design Hamiltonian is an effective and feasible way of producing pseudorandom evolution operators.

Our second problem refers to how to test randomness of the evolution operators produced in experiment. Recent theoretical studies have suggested that tools such as out-of-time-order correlators [24, 25], Rényi entanglement entropies [26], or neural networks [27] may serve as diagnostics of unitary designs. However, there is much lesser experimental study. Such a difficulty can arise from the complexity in manipulating and detecting systems at scale. For instance, in Ref. [25] it was shown that a natural probe of randomness, namely frame potential, can be expressed in terms of out-of-time-order correlators. However, these correlator functions may become difficult to estimate at late times of the design Hamiltonian evolution, because they tend to be saturated to their corresponding Haar values, which are exponentially small and can not be determined accurately from experiment. Actually, previous experimental work on the measurement of out-of-time-order functions were majorly focused on their short-time decay part [34, 42]. In our study, we are concerned about not

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only the short-time, but also the long-time behaviour of the pseudorandomness generation process, as the former features the convergence property, and the latter can signal the onset of pseudorandomness. To this end, we make use of the multiple-quantum coherence (MQC) method, a well-established technique from the realm of solid-state NMR [28, 29]. Recently, MQCs attracted great interests for their applications in studying the dynamical and statistical behaviour of complex quantum systems, such as localization-delocalization transition [30–32], buildup of multiparticle entanglement [33], and information scrambling [34]. Here, we show with experimental results that MQC spectra can also be used as a suitable means for detecting the time-development of pseudorandomness in our 12-qubit system.

**Definitions.**—We start with reviewing the definitions of random unitary matrices and unitary designs. Let \( U(d) \) denote the group of \( d \times d \) unitary matrices. Consider an ensemble of unitary operators \( \mathcal{E} = \{ U_i \} \) where \( U_i \in U(d) \). Random unitary matrices \( \mathcal{E}_{\text{Haar}} \) are the ensemble of unitary matrices uniformly distributed with respect to the Haar measure on \( U(d) \). An ensemble \( \mathcal{E} \) is said to be an approximate unitary design if it is close to the Haar ensemble \( \mathcal{E}_{\text{Haar}} \). More precisely, \( \mathcal{E} \) forms an \( \epsilon \)-approximate \( k \)-design, if for every monomial \( P(U) = U_{i_1 j_1} \cdots U_{i_n j_n} U_{m_1 n_1} \cdots U_{m_k n_k} \) of a degree not more than \( k \), its average over \( \mathcal{E} \) is \( \epsilon \)-close to that over the Haar ensemble \( \mathcal{E}_{\text{Haar}} \), i.e., \( |\langle P(U) \rangle_{\mathcal{E}} - \langle P(U) \rangle_{\mathcal{E}_{\text{Haar}}}| \leq \epsilon \) [17].

Approximate unitary designs can be realized in a number of ways, among which the design Hamiltonian approach is relatively easier to implement experimentally. A design Hamiltonian is, by definition, a physically local Hamiltonian whose interactions vary randomly at each time step and the dynamics of which forms a unitary matrix after a threshold time [24]. Put it more strictly, an \( \epsilon \)-approximate \( k \)-design Hamiltonian with \( l \)-local interaction is a random \( l \)-local Hamiltonian \( \mathcal{H} \), where there exists \( t_0 > 0 \) such that, for most of the time \( t \geq t_0 \), the propagator \( U(t) = \int_0^t \exp(-i\mathcal{H}s)ds \) generated by \( \mathcal{H} \) is an \( \epsilon \)-approximate unitary \( k \)-design. Here, the shortest such time \( t_0 \) is called the design time of \( \mathcal{H} \).

**Experimental scheme.**—In experiment, we chose the per-\(^{13}\)C-labeled dichlorocyclobutanone derivative dissolved in d6-acetone, which contains 7 labeled carbon nuclei and 5 proton nuclei and hence serves as a 12-qubit system; see Fig. 1(a). Experiment was carried on a Bruker Avance III 700 MHz spectrometer at room temperature. The system Hamiltonian under the weak coupling approximation reads

\[
\mathcal{H}_S = \sum_{i=1}^{12} \Omega_i \sigma_i^z/2 + \pi \sum_{i<j}^{12} J_{ij} \sigma_i^z \otimes \sigma_j^z / 2,
\]

where \( \Omega_i \) is the precession frequency of the spin \( i \) in rotating frame, and \( J_{ij} \) is the scalar coupling strength between spins \( i \) and \( j \); see Supplementary Material [35] for their values.

Our strategy to achieve quantum pseudorandomness here adapts the design Hamiltonian construction developed in Ref. [24]. The experimental scheme consists of applying a series of random refocusing pulse sequences generated by a design Hamiltonian, starting from the identity, and approaching randomly distributed unitaries over the whole unitary group as time passes. The trajectories represent different time-evolutions. (c) Schematic illustration of random refocusing pulse sequences that are applied to our 12-qubit system to produce random Hamiltonian evolutions. The small rectangles represent single-qubit \( \pi \) rotations.

**Results.**—In experiment, we chose the per-\(^{13}\)C-labeled dichlorocyclobutanone. (b) Intuitive picture of time-evolution operators generated by a design Hamiltonian, starting from the identity, and approaching uniformly distributed with respect to the Haar measure on \( U(d) \). More precisely, \( \mathcal{E} \) forms an \( \epsilon \)-approximate \( k \)-design, if for every monomial \( P(U) = U_{i_1 j_1} \cdots U_{i_n j_n} U_{m_1 n_1} \cdots U_{m_k n_k} \) of a degree not more than \( k \), its average over \( \mathcal{E} \) is \( \epsilon \)-close to that over the Haar ensemble \( \mathcal{E}_{\text{Haar}} \), i.e., \( |\langle P(U) \rangle_{\mathcal{E}} - \langle P(U) \rangle_{\mathcal{E}_{\text{Haar}}}| \leq \epsilon \) [17].

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Our strategy to achieve quantum pseudorandomness here adapts the design Hamiltonian construction developed in Ref. [24]. The experimental scheme consists of applying a series of random refocusing pulse sequences with change-of-basis operation \( H^\otimes n \) (\( H \) is the Hadamard transform) in between; see Fig. 1(c). Refocusing sequence is commonly used in NMR spectroscopy for adjusting effective couplings between nuclei spins. Usually it is composed of a set of single-qubit \( \pi \) pulses about \( x \) or \( y \) axis [36]. Here, by random refocusing sequence we mean that the \( \pi \) rotations therein are applied at random time. Such type of sequences has been previously shown to be useful in constituting randomized dynamical decoupling protocols with good convergence and stability [37, 38]. Now, to specify the concrete form of random refocusing sequence to be used in experiment, we fix the time length of the sequence to be \( T/2 \) and introduce a set of 8-tuple column vectors \( \lambda = \{ \lambda^{(m)} : m = 1, 2, \ldots \} \) with their entries being randomly chosen from the unit interval. For each \( m \), \( \lambda^{(m)} \) represents a random refocusing sequence composed of 8 \( \pi \) pulses, in which the \( i \)-th \( \pi \) pulse is applied on the \( i \)-th qubit at time \( \lambda^{(m)}_i T/2 \). By applying the \( m \)-th random refocusing sequence we will get dynamic evolution governed by the following effective disordered Hamiltonian

\[
\mathcal{H}^{(m)}_Z = \sum_i^{12} \Omega_{i,m}^{\text{eff}} \sigma_i^z + \sum_{i<j}^{12} J_{ij,m}^{\text{eff}} \sigma_i^z \otimes \sigma_j^z,
\]

where the coefficients \( \Omega_{i,m}^{\text{eff}} \) and \( J_{ij,m}^{\text{eff}} \) are determined by [35]

\[
\Omega_{i,m}^{\text{eff}} = (1 - 2\lambda^{(m)}_i)\Omega_i, \quad J_{ij,m}^{\text{eff}} = (1 - 2\left|\lambda^{(m)}_i - \lambda^{(m)}_j\right|)J_{ij}.
\]

Now our design Hamiltonian of the entire sequence goes: at time \( t \), let \( m = \lfloor t/(T/2) \rfloor \), then

\[
\mathcal{H}(t) = \begin{cases} \mathcal{H}^{(m)}_Z, & \text{if } m \text{ is odd;} \\ H^\otimes n \mathcal{H}^{(m)}_Z H^\otimes n, & \text{if } m \text{ is even.} \end{cases}
\]
bases into Pauli-\sigma_x bases. It is expected that the alternate approximations of time-evolutions under dual bases would quickly approach quantum pseudorandomness.

Note that our random refocusing sequences realize a design Hamiltonian of the same form as, but with a different parameter set from, the one proposed in Ref. [24]. In the original construction in [24], it was theoretically proved that \( \mathcal{H}(t) \) in Eq. (5) can generate an \( \epsilon \)-approximate unitary design within polynomial time if the coefficients are independently and uniformly distributed. Ref. [24] also pointed out that it is possible to use parameters from different sets, which could result in varied quality and efficiency of unitary design generation. Here in our construction, from Eqs. (3) and (4) we have that, the coefficients \( \Omega_{\nu,M}^{\text{eff}} \) remain uniformly distributed, but the coefficients \( J_{\nu,M}^{\text{eff}} \) are not. The main reason that we choose a different parameter set from the original scheme is due to consideration of experimental difficulty. Because there exist considerable decoherence effects in the sample, it is desirable that the pulse length in experiment be as short as possible. So our construction avoids coupled evolutions between distant spins. Besides, since the protons have relatively close resonance frequencies, which implies longer time required to control them separately, it would be better to perform collective operations on them. With these restrictions in mind, it turns out that, among others, our experimental sequence is one simplest form of random refocusing sequence that could be realized with reasonable accuracy on our molecule; see Supplementary Materials for more details [35].

We have to check to what extent our Hamiltonian forms an approximate design Hamiltonian. A useful test for unitary designs is made using the notion of frame potential [39]. Let \( \mathcal{E} = \{ U_i \} \) be an ensemble, the \( k \)-th frame potential is defined as the average of \( k \)-th powers of the ensemble elements’ Hilbert-Schmidt overlaps [40]

\[
F^{(k)}_\mathcal{E} = \frac{1}{|\mathcal{E}|^2} \sum_{i,j \in \mathcal{E}} \left| \text{Tr} \left( U_i U_j^\dagger \right) \right|^{2k},
\]

and there is \( F^{(k)}_\mathcal{E} \geq F^{(k)}_{\text{Haar}} = k! \), where equality holds iff \( \mathcal{E} \) is a \( k \)-design. Thus the deviation \( F^{(k)}_\mathcal{E} - F^{(k)}_{\text{Haar}} \) can serve as a measure of how close \( \mathcal{E} \) is to a \( k \)-design. For large-sized systems \( d \geq k \), \( \mathcal{E} \) must contain at least \( |\mathcal{E}| \geq d^{2k}/k! \) unitaries to become a \( k \)-design [41]. This implies that exact frame potential calculation is intractable. We thus have to turn to statistical estimation. Note that

\[
F^{(k)}_\mathcal{E} = \frac{d^{2k}}{|\mathcal{E}|^2} + \frac{|\mathcal{E}| (|\mathcal{E}| - 1)}{|\mathcal{E}|^2} \tilde{F}^{(k)}_\mathcal{E},
\]

where

\[
\tilde{F}^{(k)}_\mathcal{E} = \frac{1}{|\mathcal{E}|(|\mathcal{E}| - 1)} \sum_{i \neq j} \left| \text{Tr} \left( U_i U_j^\dagger \right) \right|^{2k}.
\]

So one has that, if \( d \geq k \), \( |\mathcal{E}| \geq d^{2k}/k! \) and \( \tilde{F}^{(k)}_\mathcal{E} \approx k! \). In numerical simulation, we statistically generate a sample of unitaries \( \mathcal{E} \) based on our random Hamiltonian evolutions and observe the convergence of \( \tilde{F}^{(k)}_\mathcal{E} \) with respect to sample size \( |\mathcal{E}| \). Fig. 2(c-d) show our numerical results for different periodic time \( T \), suggesting that for a range of periods and after about two rounds of evolution, the estimated frame potentials converge to their corresponding Haar values. The simulation results give strong evidences that our design Hamiltonian can generate ensemble of unitaries with significant amount of randomness.

Probing quantum pseudorandomness. We perform MQC growth experiments to detect the developed quantum pseudorandomness. An outline of the experimental procedure is shown in Fig. 3(a). Basically, the system undergoes a multiple-quantum process consisting of the following steps: (i) start from a simple operator \( \rho(0) \) (e.g., a localized state); (ii) evolve under our design Hamiltonian in Eq. (5); (iii) a collective rotational operator \( \phi_z = e^{-iM_z \phi} \) is applied, here \( M_z = \sum_i \sigma_z^i/2 \); (iv) the random evolution is reversed. We then measure the overlap of the final state with the initial state, resulting in signal

\[
S(\phi,t) = \text{Tr} \left[ e^{iHt} \phi_z e^{-iHt} \rho(0) e^{iHt} \phi_z^* e^{-iHt} \rho(0) \right] = \text{Tr} \left[ \phi_z \rho(t) \phi_z^* \rho(t) \right].
\]

Let \( \nu \) denote, for the basis \( |i\rangle \langle j| \) in the Zeeman representation, the difference between two quantum numbers: \( \nu = \langle i|M_z|i - \langle j|M_z|j \rangle \). Divide \( \rho(t) \) into blocks as \( \rho(t) = \sum_\nu \rho_\nu \) where \( \rho_\nu \) is the submatrix composed of all the order-\( \nu \) elements, and note that \( \phi_z \rho_\nu(t) \phi_z^* = e^{-i\nu \phi} \rho_\nu(t) \), there is thus

\[
S(\phi,t) = \sum_\nu e^{-i\nu \phi} I(\nu,t),
\]
where \( I(\nu, t) = \text{Tr} \left[ \rho(t)^2 \right] \). Now it is clear that the steps taken above are to ensure that in observing the multiple-quantum signal, all contributions to a given order of coherence are generated with the same phase. If we measure \( S(\phi, t) \) as a function of \( \phi \) at a fixed time \( t \) and then perform a Fourier transform with respect to \( \phi \), then we are able to extract all the amplitudes \( I(\nu, t) \) of \( \rho(t) \), which is often referred to as the MQC spectrum. Furthermore, with varying the evolution time \( t \) we will see the growth of MQCs.

What would \( I(\nu, t) \) look like typically if the evolution operator \( U(t) \) is truly random? Intuitively, under a Haar random operation, all possible coherences will be excited with equal probability. Then typically the total intensity within a given order \( \nu \) is related simply to the number of transitions consistent with that order. In an \( n \)-spin system, the number of equivalent configurations for a coherence of order \( \nu \) is \( C_{2n}^{\nu} \), which is well approximated by \( 2^{2n}(n\pi)^{-1/2}\exp(-\nu^2/n) \) for \( n > 6 \). In this picture, the resulting MQC spectrum typically shows a Gaussian pattern, i.e., \( I_{\text{typical}}(\nu) \sim \exp(-\nu^2/n) \). More details of the derivation are presented in Supplementary Materials [35]. This typical behaviour has been observed extensively in solid-state NMR where the spin dynamics is rather complex [28, 29]. Accordingly, we expect in our experiment that at long time \( t \),

\[
I(\nu, t) \rightarrow I_{\text{typical}}(\nu).
\]

Therefore, the essential idea taken here for probing the onset of quantum pseudorandomness is to measure the MQC spectrum, and then compare it with \( I_{\text{typical}}(\nu) \).

Fig. 3 shows our experimentally extracted MQC intensity distributions in our 12-qubit system at the first and second round of design Hamiltonian evolution. Here, the results are taken for \( \rho(0) = \sigma_z^\otimes 12, T = 30 \) ms, and a randomly generated array \( \lambda \) whose entries are given in Supplementary Materials [35]. The shaped pulses for implementing the random \( \pi \) rotations are obtained from the pulse compiler technique [43, 44]. The simulated fidelities of these \( \pi \) rotations as well as the Hadamard transform are all above 98.5%, with consideration of control field inhomogeneities. Pulse imperfections and decoherent effects accumulate over rounds of evolution, and the subsequent degradation in performance unavoidably reduces the signal-to-noise ratio in multiple-quantum signal observation. Importantly, while these nonideal processes deteriorate the overall fidelity of the MQC spectra, a tendency for coherences of higher order to develop with time is clearly evident in the spectra shown in Fig. 3(d,g). In particular, we put the typical MQC profile \( I_{\text{typical}}(\nu) \) in Fig. 3(g) for comparison. And we find that, the experimentally observed redistribution of spectral intensity into high-order coherences is a tangible manifestation of the growth of quantum pseudorandomness during the evolution period.
Discussions.—To generate pseudorandom operations requires the ability of making extensive control over the system’s degrees of freedom. NMR systems are well suited to study pseudorandomness generation process, featuring unique control in preparation, manipulation, and detection. Thus they make excellent testbeds to realize the ideas. Our approach to the study of random spin dynamics employs refocusing technique and multiple-quantum NMR technique. Our experimental results demonstrate the usefulness of the design Hamiltonian method in generating highly complex evolutions. In particular, there is no need to perform coupled operations between physically nonadjacent spins and no fine control of time is required. Because of the wide applicability of pseudorandom quantum operators, we expect the techniques developed and tested here will find broad applications in future quantum information protocols.

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In experiment, the reference frequencies of the $^{13}$C channel and $^1$H channel are set to be $O_1 = 20696$ Hz and $O_2 = 2696$ Hz respectively. So in the rotating frame, the system Hamiltonian takes the form

$$\mathcal{H}_S = \sum_{i = 1}^{12} \frac{\Omega_i \sigma_z^i}{2} + \pi \sum_{i < j}^{12} J_{ij} \sigma_z^i \otimes \sigma_z^j / 2,$$

where $\Omega_i$ is the precession frequency of the spin $i$, $\Omega_i = -(\omega_i - O_1)$ for $i \leq 7$ and $\Omega_i = -(\omega_i - O_2)$ for $i \geq 8$. 

The following table gives the molecular parameters including $\omega_i$ (diagonal) and $J_{ij}$ (off-diagonal).

| $J_2$ (s) | C1 | C2 | C3 | C4 | C5 | C6 | C7 | H1 | H2 | H3 | H4 | H5 |
|----------|----|----|----|----|----|----|----|----|----|----|----|----|
| 0.4      | 0.31| 0.44| 0.25| 0.25| 0.4 | 0.38| 0.29| 0.39| 0.34| 0.15| 0.30|

A critical problem in our study is to devise an experimental scheme for probing the degree of pseudorandomness generated from our design Hamiltonian evolution. Three approaches can be identified: (i) estimating frame potential of the generated evolution operators; (ii) measuring a complete set of out-of-time-order correlators (OTOCs); (iii) studying quantum coherence (MQC) technique. The first two approaches can quantitatively and completely determine to what extent a unitary ensemble forms a k-design, however, they are practically hard to realize. The last approach probes the spreading of quantum coherences over system’s degrees of freedom.
freedom. Actually, it is revealed in Ref. [33] that multipleg quantum coherences is a specific type of OTOCs. Although MQCs measurement does not offer a complete characterization of pseudorandomness, it provides rich dynamical and statistical information and is experimentally accessible.

A. Frame Potential

Frame potential is a quantity measuring the 2-norm distance between the Haar ensemble and the \( k \)-fold \( E \)-channel. For an ensemble of unitary operators \( \mathcal{E} \), the \( k \)-th frame potential is defined by the following sum

\[
F^{(k)}_\mathcal{E} = \frac{1}{|\mathcal{E}|^2} \sum_{i,j} |\text{Tr}(U_i U_j^\dagger)|^{2k},
\]

(12)

Denote the frame potential for the Haar ensemble as \( F^{(k)}_{\text{Haar}} \). Then [5]

1. \( F^{(k)}_{\text{Haar}} = k! \), which holds for \( k \leq d \).
2. For any ensemble \( \mathcal{E} \), there is \( F^{(k)}_\mathcal{E} \geq F^{(k)}_{\text{Haar}} \); here equality holds iff \( \mathcal{E} \) is \( k \)-design.

The method of characterizing a unitary ensemble in terms of frame potential is exact. However, it’s rather difficult to estimate \( F^{(k)}_\mathcal{E} \) in experiment. First, an ensemble has to contain exponential number of elements to become a design [41], it is not realistic to generate exponential number of evolution operators in experiment. Second, if we make the estimation from a feasible number of evolution operators, measuring overlaps between these evolution operators is difficult. What’s more, as the design Hamiltonian dynamics grows sufficiently random, the overlap between two random evolution operators typically gets exponentially small such that it can not be determined accurately from experiment.

B. OTOCs

Let \( \mathcal{E} = \{U_i\} \) be generated from a design Hamiltonian \( \mathcal{H}(t) \). The out-of-time-order correlator is defined as

\[
\langle AU_i U_i^\dagger AU_j U_j^\dagger \rangle_\mathcal{E},
\]

where \( A \) and \( B \) are local observables, and \( \langle \cdots \rangle_\mathcal{E} \) denotes averaging over the ensemble \( \mathcal{E} \).

Ref. [5] established the following formula

\[
F^{(k)}_\mathcal{E} = \frac{d^2}{dE^2} \sum_{A_1,\ldots,A_k,B_1,\ldots,B_k} \left| \text{Tr} \left(A_1 U_1 B_1 U_1^\dagger \cdots A_k U_k B_k U_k^\dagger \right) \right|^2,
\]

here, summations are over all possible Pauli operators. This gives that, frame potential can be expressed as a certain average of OTOC functions. Therefore, the effect of design Hamiltonian evolution on decreasing the frame potential is equivalent to that on the decay of OTOCs. Owing to this close connection, OTOC measurement could thus be used as another means of pseudorandomness detection. However, from experimental aspect of view, it can be readily seen from the above formula that, to get an exact quantification of the random dynamics an exponential number of OTOCs are involved. Furthermore, the random dynamics generated from a design Hamiltonian should quickly saturate the OTOC functions to their Haar random averages, which are exponentially small. Actually, previous OTOC measurement experiments were majorly focused on the short-time decay rather than the long-time steady behaviour of OTOC dynamics.

C. Statistics of MQC Growth Experiment

MQC growth experiments were first developed in solid-state NMR. The basic idea is that, an operator \( A(0) \) that is initially localized, e.g., on a single site of a spin network, will evolve under random evolution \( U(t) \) into a vastly more complicated operator \( A(t) = U(t) A(0) U(t)^\dagger \). The coherences of \( A \) should spread over the entire space. We can perform MQC growth experiment to get the MQC spectrum of \( A(t) \)

\[
I_\nu = \text{Tr} \left[A^{2}_\nu(t) \right],
\]

(13)

where \(-n \leq \nu \leq n\), and \( A_\nu \) is the submatrix of \( A \) composed of all the order-\( \nu \) elements. Now the question is, what would \( I_\nu \) look like typically if \( U(t) \) is random?

The MQC intensities \( I_\nu \) are actually polynomials of elements of \( U(t) \). Moments of polynomials on random unitaries can be exactly evaluated [B. Collins, Int. Math. Res. Not. 17, 953 (2003)]. Let \( A \) be a traceless and normalized Hermitian operator. If \( U \) is an \( d \times d \) Haar-distributed unitary matrix, and suppose \( d \) is large, then

\[
E \left| \langle \alpha | U A U^\dagger | \beta \rangle \right|^2 \approx \frac{|A|^{2}}{d^2} = \frac{1}{d^2}.
\]

(14)

In a system of \( n \) spins, the number of transitions with a given

FIG. 4. Typical MQC distributions when \( U \) is random, which shows an approximate Gaussian pattern.
\( \nu (-n \leq \nu \leq n) \) is given by a binomial distribution

\[
\mathcal{N}(\nu, n) = \binom{2n}{n-\nu} \frac{(2n)!}{(n+\nu)!(n-\nu)!}.
\] (15)

The MQC spectrum thus takes the form

\[
I_{\text{typical}}(\nu) \approx \frac{1}{d^2} \binom{2n}{n-\nu} \sim \exp(-\nu^2/n).
\] (16)

When \( n = 12 \), this gives the distribution shown in Table I.

### III. EXPERIMENTAL METHODS

#### A. Random \( \pi \) Pulse Sequence

Our experiment of quantum pseudorandomness generation is based on the design Hamiltonian approach. Our design Hamiltonian is given by Eq. (2-5) of the main text. Such type of design Hamiltonian can be implemented through the NMR refocusing technique. As we have described in the main text, the sequence that we use is specified by a random array \( \lambda \).

The principle that a random refocusing sequence would result in an effective Hamiltonian of the form Eq. (2-4) of the main text can be seen by just considering the simple 2-qubit case. See the following figure, where the rectangles represent \( \pi \) pulses about \( x \) (or \( y \)) axis:

\[
\begin{array}{c}
\pi \\
\lambda_1 \tau \\
\pi
\end{array}
\begin{array}{c}
\pi \\
\lambda_2 \tau \\
T
\end{array}
\]

Let \( \lambda_1, \lambda_2 \) be two random numbers, suppose \( \lambda_1 \leq \lambda_2 \), the dynamic evolution can be written as

\[
U_T = e^{-i\mathcal{H}_S \lambda_1 \tau} X_1 e^{-i\mathcal{H}_S (\lambda_2 - \lambda_1) \tau} X_2 e^{-i\mathcal{H}_S (1-\lambda_2) \tau} X_{1,2}.
\]

Note that

\[
X e^{-i\mathcal{Z}_\alpha} X = e^{i\mathcal{Z}_\alpha},
\]

\[
X_1 e^{-i\mathcal{Z}_1 \mathcal{Z}_2} X_1 = e^{i\mathcal{Z}_1 \mathcal{Z}_2},
\]

\[
X_2 e^{-i\mathcal{Z}_1 \mathcal{Z}_2} X_2 = e^{i\mathcal{Z}_1 \mathcal{Z}_2}.
\]

Substituting these formulas into \( U_T \), one can get

\[
U_T = e^{-i\mathcal{H}_{\text{eff}} \tau},
\]

### TABLE I. Typical MQC intensity distribution of a random state on a 12-spin system.

| \( \nu \) | 0  | \pm1 | \pm2 | \pm3 | \pm4 | \pm5 | \pm6 | \pm7 | \pm8 | \pm9 | \pm10 | \pm11 | \pm12 |
|----------|----|------|------|------|------|------|------|------|------|------|------|------|------|
| \( I_{\text{typical}}(\nu) \) | 0.1612 | 0.1488 | 0.1169 | 0.0779 | 0.0206 | 0.0080 | 0.0025 | 0.0006 | 0.0001 | 1.65e-5 | 1.43e-6 | 5.96e-8 |

#### B. Pulse Design and Optimization

Pulse design and optimization for implementing the random \( \pi \) sequences and their reverses on a 12-spin system is not an easy task. We have to combine a number of pulse techniques together to achieve the goal. On the whole, we execute the following step by step:

1. Construct an approximate circuit that realizes the target evolutions approximately but is as simple as it can be.

2. Use selective pulses to implement the single-qubit rotations in the approximate circuit. Here, we use a pulse sequence compilation program to reduce the phase errors of selective pulse control. The resulting selective pulse sequence serves as a good initial pulse for further gradient-based optimization.

3. Use subsystem-based pulse optimization algorithm to further increase the pulse control fidelity.

The procedure is illustrated in Fig. 5.

![Schematic of pulse design and optimization for design Hamiltonian evolution (left) and its reverse (right).](image)
1. Circuit and Initial Pulse Construction

The first step of pulse optimization is to construct an initial pulse, either from a random guess or through specific design, which serves as the starting point for subsequent optimization. A suitably constructed initial pulse makes the optimization procedure easier to reach a final pulse with satisfying accuracy. According to our practice experiences, this is especially important for systems with number of qubits more than seven.

Our strategy for constructing an initial pulse for random $\pi$ sequences and their reverses is to design approximate circuits in terms of an approximate Hamiltonian. Concretely, we consider a simplified coupling network in which we ignore the small couplings and the small differences between large couplings of the original Hamiltonian. Such simplification manifests which couplings should be majorly accounted for in order to accomplish reversed evolutions of $\mathcal{H}_Z^{(m)}$, and thus enables direct circuit construction. The circuits thus constructed, if we turn back to the real Hamiltonian, generate evolutions that deviate the corresponding desired ones slightly, thus provide good starting points for further optimization.

Now we describe the strategy in more details. Consider a refocusing operation

$$ R_y^{1357}(\pi) = R_y^1(\pi)R_y^3(\pi)R_y^5(\pi)R_y^7(\pi), $$

notice that

$$ U_{-\mathcal{H}_Z^{(m)}}(t) = R_y^{1357}(\pi)U_{\mathcal{H}_Z^{(m)}}(t)R_y^{1357}(\pi) \cdot U_{ZZ}^{\text{ref}}(t)U_{ZZ}^{\text{ref}}(t), $$

where $U_{ZZ}^{\text{ref}}(t)$ and $U_{ZZ}^{\text{ref}}(t)$ are $Z$- and $ZZ$-type evolution errors coming from those evolutions that are not refocused by $R_y^{1357}(\pi)$, respectively. Here, $Z$-type error terms are easy to handle with. The point is that, whenever there is a rotation about $z$ say $R_z(\gamma)$: (i) if it is followed by a period of free evolution, their order can be interchanged; (ii) if it is followed by a reverse rotation $R_z(\theta)$, it can be moved across that rotation according to: $R_z(\theta)R_z(\gamma) = R_z(\gamma)R_z(-\theta)$. Therefore, $Z$-type errors, whenever encountered, actually need not be executed and can always be moved one step forward till the end of the circuit [36, 43, 44]. The $ZZ$-type error terms are mainly due to unrefocused couplings $\{J_{ij}\} \setminus \{J_{ij} : \text{only one of } i, j \in \{1, 3, 5, 7\}\}$. From the parameter table, it is obviously seen that the unrefocused couplings are mostly small, except for $J_{57}$, $J_{2,11}$ and $J_{48}$. Summarizing these observations, we expect that the circuits

$$ R_y^{1357}(\pi)U_{\mathcal{H}_Z^{(m)}}(t)R_y^{1357}(\pi) $$

are good candidates on which we seek to achieve $U_{-\mathcal{H}_Z^{(m)}}(t)$ through pulse optimization.

Another benefit of the above strategy is that, because we have ignored small couplings, the resulting circuits could be much shorter than those if we do in other ways.

2. Selective Pulse Sequence Compilation

The (approximate) circuit for each $\mathcal{H}_Z^{(m)}$ (or $-\mathcal{H}_Z^{(m)}$) evolution is composed of free evolutions and 8 $\pi$ rotational gates.

To realize the rotational gates, we use frequency selective pulses. For example, a rotational gate on a specific spin can be realized by a rotating Gaussian that is in resonance with that spin. In order that the number of control parameters after pulse discretization be as few as possible, we adopt relatively large time step length $\tau = 20 \mu s$.

It is important to be aware of that a selective pulse just approximately implements the target operation. Various types of errors arise when transferring a circuit directly into a selective pulse sequence without correction. What’s more, as the number of gates contained in the circuit grows large, the error accumulation will become increasingly serious. To address this problem, we use the pulse sequence compilation program developed in Refs. [43, 44]. The compilation program systematically adjusts the pulse parameters of an arbitrary input selective pulse sequence so that errors up to first-order can be corrected. The compilation procedure is efficient. With application of the compilation method to our pulse sequence, the control accuracy is greatly improved. Although the compilation program can not eliminate all control imperfections that higher-order errors still exist, it is still quite useful since that, the pulse sequence after compilation is of relatively high fidelity and can be used as a good starting point for subsequent gradient-based optimization.

3. Subsystem-based GRAPE

Gradient ascent pulse engineering (GRAPE) is a numerical algorithm widely used for optimal control pulse search in quantum control. However, it is challenging to run GRAPE for as large as a 12-qubit system, due to that this involves computations of $2^{12}$-dimensional matrix multiplications and exponentials that require substantial amount of memory and time cost. A variant of GRAPE, namely subsystem-based GRAPE (SSGRAPE) can reduce the computational cost required to some extent [43]. SSGRAPE works, for our 12-qubit system, as follows. We divide the whole system into two subsystems, $S_A = \{C_1, C_2, C_3, H_2, H_4\}$ and $S_B = \{C_4, C_5, C_6, C_7, H_1, H_5\}$, each consisting of 6 spins. The only large couplings between these two subsystems is $J_{C_2C_7}$, so they can be approximately viewed as isolated. Their respective Hamiltonian, $\mathcal{H}_{S_A}$ and $\mathcal{H}_{S_B}$, can be obtained from $\mathcal{H}_S$ by tracing the other subsystem. Suppose we intend to find a pulse to implement a target operation $U$ (e.g., single-qubit rotation) of the form $U = U_{S_A} \otimes U_{S_B}$. Instead of searching on the whole system, we require the pulse to be optimized should realize desired subsystem operations on both subsystems. Let $\mathcal{H}_C(t)$ denote the time-dependent control Hamiltonian, let

$$ V(t) = \int_0^t \exp (-i(\mathcal{H}_S + \mathcal{H}_C(s))) , $$

$$ V_{S_A}(t) = \int_0^t \exp (-i(\mathcal{H}_{S_A} + \mathcal{H}_C(s))) , $$

$$ V_{S_B}(t) = \int_0^t \exp (-i(\mathcal{H}_{S_B} + \mathcal{H}_C(s))) . $$
The overall fitness function is \( f = \langle \text{Tr}(U^\dagger V(t))^2 \rangle \), while in SSGRAPE we attempt to maximize the fitness function

\[
f_S = \left( \langle \text{Tr}(U_S^† V_S(t))^2 \rangle + \langle \text{Tr}(U_B^† V_B(t))^2 \rangle \right) / 2, \tag{18}\]

It is expected that when \( f_S \) is sufficiently high, then on the whole system, \( V(t) \) will get close to the target \( U \). In this sense, the 12-qubit GRAPE optimization problem is approximately treated as a two-GRAPE optimal pulse control problems.

### IV. SOME SIMULATION RESULTS

In Fig. 2 of the main text, we showed that, via frame potential estimations, our design Hamiltonian can generate approximate unitary 2-designs. Here, before we present our experimental results, we give additional MQC simulation results that reveal more details of the onset of pseudorandomness.

To characterize the relative deviation between the MQC spectra with respect to the typical MQC distribution, we use the following quantities:

\[
e(t) = \frac{||I_{\rho}(t = 2T) - I_{\text{typical}}||}{I_{\text{typical}}} \tag{19}\]

In Table. II, we made calculations on \( e(t = 2T) \) for MQCs at the second round of design Hamiltonian evolution, where the input initial operators \( \rho_i \) are randomly selected from the Pauli group, and we have randomly created two different focusing matrices. It is found that, for the tested examples, the relative distance \( e \) rarely exceeds 3%, which clearly indicates that under our design Hamiltonian evolution, the long-time MQC spectrum gets close to the typical distribution.

To see the MQC transient behaviour, we made further com-
FIG. 6. Simulated MQC spectra results corresponding to the states at \( t = 0, T/2, T, 3T/2, 2T \) (\( T = 30 \) ms), starting from the following initial operators: (a) \( \sigma_z^0 \sigma_z^0 \), (b) \( \sigma_x^0 \sigma_x^0 \), (c) \( \sigma_z^0 \sigma_x^0 \), (d) \( \sigma_z^0 \sigma_y^0 \), (e) \( \sigma_z^0 \sigma_z^0 \). Here, the refocusing matrix \( \lambda \) is randomly generated as Eq. (20).

FIG. 7. Simulated results showing more detailed transient behaviour of MQC spectra corresponding to the states evolving at \( T/2 \leq t \leq T \) (\( T = 30 \) ms), starting from \( \rho_i = \sigma_z^2 \sigma_z^2 \). Here, the refocusing matrix \( \lambda \) is randomly generated as Eq. (20).

dulations on the MQC dynamic evolution, which are shown in Fig. 6 and Fig. 7. The refocusing matrix is randomly generated as follows

\[
\lambda = \begin{pmatrix}
0.9494 & 0.0635 & 0.8321 & 0.4605 \\
0.2564 & 0.3735 & 0.7538 & 0.6455 \\
0.9899 & 0.1663 & 0.6219 & 0.5135 \\
0.3498 & 0.2313 & 0.3941 & 0.8144 \\
0.2085 & 0.0522 & 0.3593 & 0.0972 \\
0.6658 & 0.9018 & 0.0889 & 0.4637 \\
0.9733 & 0.7933 & 0.3417 & 0.5898 \\
0.6227 & 0.373 & 0.5487 & 0.1872
\end{pmatrix}
\] (20)
Here, we randomly selected a set of local Pauli basis elements as initial states, so that we can see the spreading of coherences. The results are in Fig. 6. Notice that our design Hamiltonian $H(t)$ (Eq. (2-5) of the main text) is composed of $H_Z$ and $H_X$, and that $H_Z$ does not change the MQC distribution, i.e., $H_Z$ just affects the distribution of the density elements within the same order coherence subspace, so actually we only detect time varying MQC spectra during the $H_X$ evolutions. Understanding this, we display in Fig. 7 the detailed MQC evolution during the second stage of the first round $T/2 \leq t < T$. To summarize, the MQC growth experiments can be used as a means for detecting system’s coherence distribution, whose temporal and long-time limiting behaviour are in accordance with that the degree of pseudorandomness grows under the design Hamiltonian evolution.

V. EXPERIMENTAL RESULTS

We have performed two experiments observing MQC growth under evolution by random refocusing sequences with different parameter sets to demonstrate the process of generating quantum pseudorandomness through design Hamiltonian evolution. The MQC growth experiment measures the following multiple-quantum signal

$$S(\phi, t) = Tr \left[ e^{iHt} \phi_z e^{-iHt} \rho(0) e^{iHt} \phi_z e^{-iHt} \rho(0) \right].$$ (21)

Here, the design Hamiltonian $\mathcal{H}$ is specified by $\lambda$. The experimental results are presented as follows.

A. Experiment I

Initial state $\rho_0 = \sigma_z^7$. The state is prepared through first destroying all polarizations except that of $H_5$ and then applying a SWAP gate $\text{SWAP}_{C_7 H_5}$ (8ms, 0.9883) to transfer the polarization to $C_7$

$$\rho_{eq} \rightarrow \sigma_z^{12} \xrightarrow{\text{SWAP}_{C_7 H_5}} \sigma_z^7.$$
The matrix \( \lambda \) is randomly generated as below:

\[
\lambda = \begin{pmatrix}
0.2710 & 0.3219 & 0.8206 & 0.3628 \\
0.7585 & 0.7204 & 0.6633 & 0.7545 \\
0.6796 & 0.8401 & 0.6154 & 0.9943 \\
0.7590 & 0.7336 & 0.6464 & 0.7608 \\
0.9323 & 0.3947 & 0.9191 & 0.2523 \\
0.3520 & 0.3888 & 0.3666 & 0.7918 \\
0.4772 & 0.8511 & 0.4400 & 0.6621 \\
0.2982 & 0.2354 & 0.1435 & 0.2457
\end{pmatrix}.
\]

The corresponding experimental pulses for implementing 1st round (34 ms, 0.9910) and 2nd round (68 ms, 0.9642) design Hamiltonian evolution are shown in Fig. 8(a). Fig. 8(b-c) show the experimental results.

**B. Experiment II**

Initial state \( \rho_0 = \sigma_1^4 \sigma_5^5 \). The state is prepared through first destroying all polarizations except that of \( H_1 \), then applying a SWAP gate \( \text{SWAP}_{C_{14}H_1} \) (10 ms, 0.9872) to transfer the polarization to \( C_4 \), and finally evolving the state under \( J_{45} \):

\[
\rho_{eq} \rightarrow \sigma_z^8 \text{SWAP}_{C_{14}H_1} \rightarrow \sigma_z^4 \frac{R_z}{2}(\pi/2) \rightarrow \sigma_y^4 \rightarrow \sigma_z^4 \sigma_5^5.
\]

The matrix \( \lambda \) is randomly generated as below:

\[
\lambda = \begin{pmatrix}
0.3992 & 0.9113 & 0.7843 & 0.6434 \\
0.1547 & 0.6535 & 0.6132 & 0.8538 \\
0.6927 & 0.7988 & 0.7138 & 0.9148 \\
0.2291 & 0.6864 & 0.6698 & 0.7613 \\
0.5762 & 0.2014 & 0.3898 & 0.7146 \\
0.4427 & 0.5866 & 0.9014 & 0.3416 \\
0.1212 & 0.7092 & 0.8785 & 0.6002 \\
0.6999 & 0.1389 & 0.1223 & 0.3085
\end{pmatrix}.
\]

The corresponding experimental pulses for implementing 1st round (34 ms, 0.9894) and 2nd round (68 ms, 0.9448) design Hamiltonian evolution are shown in Fig. 9(a). Fig. 9(b-c) show the experimental results.