Maximal entanglement between a quantum walker and her quantum coin for the third step and beyond regardless of the initial state

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We study maximal entanglement generation between a walker and her coin via a discrete-time quantum walk, in which the coin operation is randomly selected from one of two coin operators set at each step. We solve maximal-entanglement generation as an optimization problem with quantum process fidelity as the cost function. Then we determine an appropriate pair of one-parameter coins along with coin sequences that generate maximal entanglement, which is available for any step beyond the second regardless of initial condition. We further simplify the coin set to comprising Hadamard and identity operations, which are feasible experimentally. Experimentally, we demonstrate a ten-step quantum walk with such coin sequences and thereby show the desired high-dimensional bipartite entanglement.

A quantum walk (QW) is the quantum version of a classical random walk [1, 2]. Due to the principle of superposition in quantum mechanics, a QW gives rise to impressive applications in quantum information science, from quantum computing [3–6] to quantum simulation [7], and from implementing quantum measurement [8–10] to exploring topological phases [11–18]. For the discrete-time QW (DTQW), entanglement can be generated between coin (c) and walker (w) position degrees of freedom of the walker [19], which is a key resource for quantum information processing [20]. Particularly, qubit-qudit entanglement generated in DTQW provides richer contents beyond qubit-qubit entanglement, such as nonlocality [21, 22], entanglement detection [23–25], entanglement of formation [26–28], survival of entanglement [29, 30], separability [31, 32], steering [33], concurrence [27, 34, 35] and discord [36–38]. The DTQW is particularly valuable for maximal entanglement generation (MEG), and our goal is guaranteed MEG after just a few steps.

In a dimensional (1D) DTQW with static coin operations (unchanging coin operation during evolution), entanglement generation depends on the initial coin state and cannot reach a maximal value [39]. Counterintuitively, by introducing disorder into the DTQW [40], e.g., randomly choosing SU(2) coin operation

\[ \hat{C}(\xi, \gamma, \zeta) = \left( \begin{array}{cc} e^{i\xi} \cos \gamma & e^{i\xi} \sin \gamma \\ e^{-i\xi} \sin \gamma & -e^{-i\xi} \cos \gamma \end{array} \right), 4\gamma, \xi, \zeta \in [0, 2\pi], \]

at each step, generated entanglement is significantly enhanced and achieves MEG asymptotically independent of initial conditions [41, 42]. Motivated by robust entanglement generation under experimental conditions with imperfections and disorder, random-coin DTQWs have been studied for various disorder configurations [43–46] and been observed experimentally in a 20-step optical network [47].

The main obstacle to utilizing a disordered DTQW for MEG is that many steps are required to reach asymptotic maximal entanglement [41], which is problematic for current experimental technologies. Alternative strategies in optimising coin-operation sequence during the evolution have been proposed aiming at MEG for few steps. Universal and optimal coin sequences are proposed to generated highly entangled states for fewer than 10 steps [48, 49]. However, the universal sequence works for an odd number of steps and for the states with vanishing relative phase, whereas the optimal sequence depends on the initial coin states. Parrondo sequences have been proposed to generate maximal entanglement at steps \( T = 3 \) and \( T = 5 \), with the expectation that many steps are needed to for MEG [50]. Recently, MEG has been realized with a position-inhomogeneous quantum walk [51]. Ideal MEG via a DTQW should work for any step number and independent of initial conditions, but previous experiments only achieve either one or the other, not both; we achieve both simultaneously for all steps beyond the second by solving an optimisation problem. Experimentally, we demonstrate the DTQW with requisite coin sequences up to 10 steps with linear optics and observe significant enhancement of entanglement generation compared to other entanglement-generation schemes.

The 1D DTQW coin-walker Hilbert space is \( \mathcal{H} = \mathcal{H}_c \otimes \mathcal{H}_w \) with

\[ \mathcal{H}_c = \text{span}\{ |0\rangle_c, |1\rangle_c \}, \mathcal{H}_w = \text{span}\{ |x\rangle; x \in \mathbb{Z} \} \]

The walker is initially localized position, \( |0\rangle_w \), with arbitrary initial coin \( |\theta, \phi\rangle_c^{in} = \cos(\theta/2)|0\rangle_c + e^{i\phi} \sin(\theta/2)|1\rangle_c \), where \( 2\theta, \phi \in [0, 2\pi] \). At step \( t \), coin operator \( \hat{C}_t \) is applied. Then the walker moves left or right entangled with the coin represented by the conditional shift operator

\[ \hat{S} = |0\rangle_c \langle 0| \otimes \sum_x |x+1\rangle_w \langle x|_c + |1\rangle_c \langle 1| \otimes \sum_x |x-1\rangle_w \langle x|, \]
Sex Sequence of coin operations
\( C_T \)
\[ \rho^{(b)} = \sqrt{\theta, \phi}^{(b)}(\theta, \phi) \]

(a) (b)

[Diagram]

Sequence of coin operations \( C_T \)
\[ \rho^{(b)} = \mathcal{E}_c(\rho^{(b)}) = |1/2 \]

for \( \mathcal{I} = \sum |x\rangle\langle x| \) the identity operator on \( \mathcal{H}_w \), and \( f \) short for ‘final’. The sequence \( C_T := \left( \hat{C}_t \right)_{t \in [T]} \) describes coin operations applied to the walker. As varying coin parameters \( \xi \) and \( \zeta \) is equivalent to varying \( \phi \) of initial state \( |\theta, \phi\rangle^{in} \) \([52]\], we replace \( \hat{C}(\gamma) \rightarrow \hat{C}(0, \gamma, 0) \). Furthermore, we restrict construction of \( C_T \) by allowing only two coin operations: \( \gamma_{0,1} \) labelled by one bit with values 0 and 1. The sequence \( C_T \) is then expressed as a bit string \( b \in \{0, 1\}^T \) of length \( T \) with 0 labelling \( C(\gamma_0) \) and 1 labelling \( C(\gamma_1) \).

Achieving walker-coin MEG at step \( T \) regardless of \( |\theta, \phi\rangle^{in} \) corresponds to designing \( C_T \) that maps any \( |\theta, \phi\rangle^{in} \otimes |0\rangle_w \) to maximally entangled walker-coin state \( |\theta, \phi\rangle_T \). Entanglement of \( |\theta, \phi\rangle_T \) is quantified by entropy

\[ S_E(|\theta, \phi\rangle_T) = -\text{tr}(\rho^{(b)}_T \log_2 \rho^{(b)}_T) = -\sum_{\lambda} \lambda \log_2 \lambda \]

of the reduced coin state \([39, 53]\) \( \rho^{(b)}_T := \text{tr}_w(|\theta, \phi\rangle_T \langle \theta, \phi|) \) and \( \lambda \) are the eigenvalues of \( \rho^{(b)}_T \). Note that \( 0 \leq S_E \leq 1 \), and \( S_E = 0 \) for separable states and 1 for maximally entangled states.

Thus, MEG evolution (3) yields maximally entangled \( |\theta, \phi\rangle_T \), which is equivalent to \( \mathcal{E}_{C_T} \left( \rho^{(in)} = |\theta, \phi\rangle^{in} \langle \theta, \phi| \right) = \frac{1}{2} \) in \( \mathcal{H}_c \), where \( \mathcal{E}_{C_T} \) is a completely-positive linear map determined by \( C_T \). A geometric illustration of \( \mathcal{E}_{C_T} \) for MEG is in Fig. 1(a), which is the depolarizing channel \( \mathcal{E}_{DP} \left( \rho^{(in)} \right) = (1 - \eta)\rho^{(in)} + \eta \frac{1}{2} \) with \( \eta = 1 \) \([54]\). Process fidelity \( F_{C_T} := \text{tr} \left( \sqrt{\sqrt{C_T} \chi_{DP} \sqrt{C_T}} \right)^2 \), is our figure of merit to design \( C_T \), where \( \chi_{DP} \) is the Pauli-matrix representation of quantum channel \( \mathcal{E}_{C_T} \). Note that \( F_{C_T} = 1 \) indicates MEG at step \( T \) regardless of \( |\theta, \phi\rangle^{in} \), and we refer to the corresponding coin sequence \( C_T \) as the optimal coin sequence.

Three parameters determine \( C_T \): \( \gamma_{0,1} \) and corresponding \( b \). We address designing optimal \( C_T \) as an optimization problem. For fixed \( T \), optimizing \( b \) and \( \gamma_{0,1} \) is achieved by minimising the cost function \( 1 - F_{C_T} \), in our case using an annealing algorithm. Results of optimisation of \( \gamma_{0,1} \) at \( T \in \{5, 10, 20\} \) are shown in Fig. 1(b) (\( b \) is not shown here). Evidently, the minimal \( 1 - F_{C_T} \) is obtained for two coin sets: \( \hat{C}(0), \hat{C}(\pi/4) \) and \( \hat{C}(\pi/2), \hat{C}(\pi/4) \). Note that \( \hat{C}(0) = \hat{\sigma}_z, \hat{C}(\pi/4) = \hat{H} \) and \( \hat{C}(\pi/2) = \hat{\sigma}_x \). The evolution unitary operator with coin operator \( \hat{\sigma}_z \), i.e., \( \hat{U} = \hat{S}_z \), makes the components \( |0\rangle_c \) and \( |1\rangle_c \) propagate in the opposite direction without interference, which has the similar effect of \( \hat{U} = \hat{S}_z \) with identity operator \( \mathcal{I} \) yielding a phase \( e^{i\pi} \) on component \( |1\rangle_c \).

We conjecture that the coin set \( \{\hat{H}, \mathcal{I}\} \) with optimized \( b \) leads to MGE as well. Along this spirit, we fix \( T \) and coin set \( \{\hat{H}, \mathcal{I}\} \) and optimize \( b \) with cost function \( 1 - F_{C_T} \). The results of \( F_{C_T} \), with \( T \) up to 20 are shown with blue dots in Fig. 1(c), in which we observe that \( F_{C_T} = 1 \) since step \( T = 3 \). To give a comparison, we also show the maximum \( F_{C_T} \) over \( 2^T \) possible \( b \) with coin sets \( \{\hat{H}, \hat{\sigma}_z\}, \{\hat{H}, \hat{\sigma}_x\}, \{\hat{H}, \hat{F}\} \) and \( \{\hat{H}\} \), where
\[ \hat{F} = [1, i, i, 1]/\sqrt{2}. \] The results with coin set \( \{\hat{H}, \hat{\sigma}_y\} \) is as same as that with \( \{\hat{H}, \hat{\mathbb{1}}\} \). For the coin set \( \{\hat{H}, \hat{\sigma}_z\} \), \( \mathcal{F}_{CT} = 1 \) is achieved at step \( T = 5 \) and \( T \geq 7 \). Asymptomatic behavior is observed with coin set \( \{\hat{H}, \hat{F}\} \) and oscillating behavior is observed in Hadamard walk.

We note that optimal \( b \) at step \( T \) is not unique. For instance, we obtain 1104 optimal \( b \) at \( T = 20 \), and we list 50 among them in Fig. 1(d). There are no obvious features of regularities and generalities of these optimal \( b \). An optimal \( b \) containing \( \{\hat{H}\} \) as less as possible is preferred in experiment, so we choose the optimal \( b \) containing one 0 or two 0s in our realization (The explicit form of \( b \) and its corresponding proof are in Appendix A).

The experimental setup to implement 1D DTQW is shown in Fig. 2(a). The coin state is encoded in the photon’s polarization degree of freedom by \( |H(V)\rangle = |0(1)\rangle \), where \( |H(V)\rangle \) denotes the horizontal (vertical) polarization. The position state is encoded in the photon’s spatial degree of freedom, i.e., the longitudinal spatial modes. Two photons in state \( |H(V)\rangle \) with central wavelength at 810 nm are generated from a periodically poled potassium titanyl phosphate (PPKTP) crystal pumped by an ultraviolet CW laser diode with the central wavelength at 405 nm. The two photons are then separated by a polarizing beam splitter (PBS), which transmits the horizontal polarization and reflects vertical polarization. The reflected photon is detected by single-photon detector (SPD) to serve as a trigger. The transmitted photon is sent into the photonic network consisting of waveplates and birefringent calcite beam displacers (BDs), in which the longitudinal spatial mode of injected photon is denoted as the start position of the walker \( |0\rangle_w \). The coin operations \( \hat{C}_t \) are realized by waveplates which rotates the polarization of photon, and the BD transmits the vertical polarization while deviating from the horizontal polarization so that BD acts as shift operation \( \hat{S} \).

To reconstruct the process matrix \( \chi_{CT}^{\exp} \), we prepare four states as initial coin states \( |\theta, \phi\rangle^\text{in}_{\psi_c} \), i.e., \( |H\rangle, |V\rangle, |+\rangle \) and \( |L\rangle \). For each step \( t \), we set the the optimal coin sequence \( \hat{C}_T \) accordingly, and reconstruct \( \rho_t^E \) using quantum state tomographic technology [54]. The experimental results of \( \mathcal{F}_{CT} \) with coin set \( \{\hat{H}, \hat{I}\} \) are shown with blue circles in Fig. 3(a). We observe that the average \( \mathcal{F}_{CT} \) from \( T = 3 \) to \( T = 10 \) is 0.9954 ± 0.0008, which is much better than the results with coin set \( \{\hat{H}, \hat{F}\} \) as shown with red squares. For the Hadamard QW, \( \mathcal{F}_{CT} < 0.8 \) and oscillates as \( T \) increases (shown with yellow triangles). We calculate the average entanglement \( \langle E \rangle \) over 296 initial coin states with the reconstructed \( \chi_{CT}^{\exp} \), and the results are shown in Fig. 3(b). The error bar indicates initial-state-independence, and we observe a stronger initial-state-independence with coin set \( \{\hat{H}, \hat{I}\} \) than the other two. This is also reflected by the geometric interpretations of \( \mathcal{F}_{CT} \) as shown in Fig. 3(c) (coin set \( \{\hat{H}, \hat{I}\} \) and Fig. 3(d) (coin set \( \{\hat{H}, \hat{F}\} \)) at \( T = 4, 6, 8 \) and 10, respectively. It is obviously that the results with \( \{\hat{H}, \hat{I}\} \) is much more dense than that with \( \{\hat{H}, \hat{F}\} \), which indicates the entanglement generation with coin set \( \{\hat{H}, \hat{I}\} \) has stronger independence of initial coin states than that with the other two. More details are shown in Appendix B.

Besides \( \mathcal{F}_{CT} \), we investigate spreading over the walker Hilbert space \( \mathcal{H}_w \). Spread of the probability distribution \( P(x,T) \) can be characterized by the normalized Shannon entropy

\[ S_S(T) = -\frac{\sum_x P(x,T) \ln P(x,T)}{\ln(T+1)}, \]

with \( \frac{1}{\ln(T+1)} \) the normalisation parameter. The walker is able to occupy \( T + 1 \) positions after \( t \) steps so that the maximal value of \( -\sum_x P(x,T) \ln P(x,T) \) is \( \ln(T+1) \), which corresponds to the uniform distribution over \( T + 1 \) positions [55]. Larger \( S_S(T) \) implies \( P(x,T) \) is more uniform. For a \( T \)-step QW associated with corresponding optimal \( C_T \), we measure the probability distribution \( P(x,T) \) at step \( T \), according to which we calculate the normalized Shannon’s entropy \( S_S(T) \). The results of \( P(x,T) \) with initial coin state \( |H\rangle \) and \( |L\rangle \) are shown in Fig. 4(a,c), and the corresponding \( S_S(T) \) are shown in Fig. 4(b,d) respectively. Compared with the other
two cases, the uniformity of QW with coin set \{\hat{H}, \hat{1}\} shows enhancement at \(T = 3\) and \(T = 7\). We also analyze the trend of probability distributions with different coin sets. More experimental results and analysis can be found in Appendix C.

In conclusion, we design coin sequences that can rigorously generate maximal entanglement between coin and her walker in 1D DTQW with three key features, that are: to be available at any \(T \geq 3\), to be independent of initial coin state and to be the simplest for experimental implementation. A comparison of our coin sequence \(C_T\) with the other coin sequences is shown in Appendix D, and MEG with our coin sequence significantly outperforms all other proposed coin sequences in three features mentioned above.

Experimentally, we realize a 10-step 1D DTQW with an optical network, and observe the MEG with our proposed coin sequence as well as other coin sequences. The results show a significant enhancement of MEG with our coin sequences, which benefits the intermediate quantum information processing that requires maximal qudit entanglement. Moreover, for few steps, the spread of probability distributions with our coin sequence is more uniform, which is favorable and useful in various quantum algorithms and quantum simulation of biological processes [55–57]. Our protocol can also be generalized to \(p\)-diluted disorder QW [58, 59], in which transport behavior can be engineered by controlling probability of coin operations.

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FIG. 4. Measured $\mathcal{P}(x, T)$ with initial coin state (a) $|H\rangle$ and (c) $|L\rangle$, according to which the calculated $S_s(T)$ are shown in (b) and (d), respectively.

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in Eq. (2) can be expressed in momentum space as

\[
\hat{S}_m = (e^{-ik}|0\rangle\langle 0| + e^{ik}|1\rangle\langle 1|) \otimes |k\rangle\langle k|
\]

Accordingly, the evolution unitary operators \( \hat{U} = \hat{S}(\hat{C} \otimes \mathbb{1}_w) \) is expressed by

\[
\hat{U}_m = \hat{S}_m(\hat{C} \otimes \mathbb{1}_w) = \left( \begin{array}{cc} e^{-ik} & 0 \\ 0 & e^{ik} \end{array} \right) \hat{C} \otimes |k\rangle\langle k| \sum_x |x\rangle\langle x| 
\]

\[
= \frac{1}{4\pi^2} \int dk \int dk' \left( \begin{array}{cc} e^{-ik} & 0 \\ 0 & e^{ik} \end{array} \right) \hat{C} \otimes \sum_x e^{-i(k-k')x}|k\rangle\langle k'| 
\]

\[
= \frac{1}{2\pi} \int dk \left( \begin{array}{cc} e^{-ik} & 0 \\ 0 & e^{ik} \end{array} \right) \hat{C} \otimes |k\rangle\langle k| 
\]

\[
= \int \hat{C}_m \otimes \frac{dk}{2\pi}|k\rangle\langle k|,
\]

where we have used the orthonormalization relation

\[
\sum_{k \in \mathbb{Z}} e^{-i(k-k')x} = 2\pi \delta(k-k'),
\]

and denote \( \left( \begin{array}{cc} e^{-ik} & 0 \\ 0 & e^{ik} \end{array} \right) \hat{C} \) as \( \hat{C}_m \). Specifically, for the coin operation \( \hat{C} = \hat{H} \) and \( \hat{C} = 1 \), we obtain

\[
\hat{H}_m = \frac{1}{\sqrt{2}} \left( \begin{array}{cc} e^{-ik} & e^{ik} \\ e^{ik} & -e^{ik} \end{array} \right), \hat{C}_m = \frac{1}{\sqrt{2}} \left( \begin{array}{cc} e^{-ik} & 0 \\ 0 & e^{ik} \end{array} \right).
\]

Thus, the dynamic evolution of initial state \( |\theta, \phi\rangle_c^{in} \otimes |0\rangle_w \) is

\[
|\theta, \phi\rangle_t = \int \prod_{t=1}^T \hat{C}_{m,t} \otimes \frac{dk}{2\pi}|k\rangle\langle k| \cdot |\theta, \phi\rangle_c^{in} \otimes \frac{dk'}{2\pi}|k'\rangle\langle k'|
\]

\[
= \int \prod_{t=1}^T \hat{C}_{m,t} |\theta, \phi\rangle_c^{in} \otimes \frac{dk}{2\pi}|k\rangle.
\]

The reduced density matrix of the coin is

\[
\rho_c^t = \text{tr}_w (|\theta, \phi\rangle_1 \langle \theta, \phi|) \quad (A7)
\]

\[
= \int \int \prod_{t=1}^T \hat{C}_{m,t} |\theta, \phi\rangle_c^{in} \langle \theta, \phi| \hat{C}_{m,t} \frac{dkdk'}{4\pi^2} |x\rangle\langle x| |k'\rangle\langle k'|
\]

\[
= \int \frac{dk}{2\pi} \prod_{t=1}^T \hat{C}_{m,t} |\theta, \phi\rangle_c^{in} \langle \theta, \phi| \hat{C}_{m,t}^\dagger
\]

2. Superoperator

We describe the evolution of density matrix of the coin by a superoperator \( \hat{L} \)

\[
\rho_c^{t+1} = \hat{L} \rho_c^t.
\]

According to Eq. (A7), the one step evolution of density matrix of the coin is

\[
\rho_c^{t+1} = \int \frac{dk}{2\pi} \hat{C}_m \rho_c^t \hat{C}_m^\dagger
\]
An arbitrary coin state $\rho_c^t$ can be described with Pauli matrices $\{1, \sigma_x, \sigma_y, \sigma_z\}$ by
\[
\rho_c^t = \frac{\alpha_0 + \alpha_3}{2} \begin{pmatrix} \alpha_0 + \alpha_3 & \alpha_1 - i\alpha_2 \\ \alpha_1 + i\alpha_2 & \alpha_0 - \alpha_3 \end{pmatrix}.
\]
(11)
where $\alpha_0 = \frac{1}{2}$ satisfying $\text{tr}(\rho) = 2\alpha_0 = 1$. Using the affine map approach [61], we represent $\hat{L}$ as a matrix acting on the $2 \times 2 \rho_c^t$, which can be further expressed as a four-dimensional column vector $\alpha_c^t$ [62]
\[
\alpha_c^t = \begin{pmatrix} \frac{1}{2} \\ \alpha_1 \\ \alpha_2 \\ \alpha_3 \end{pmatrix}.
\]
(12)
where $\alpha_i = \frac{1}{2} \text{tr}(\rho \sigma_i)$. Along this spirit, dynamic evolution of density matrix of the coin can be expressed as
\[
\alpha_c^t = \int \frac{dk}{2\pi} \prod_{t=1}^T \hat{L}_t \alpha_c^{in},
\]
(13)
where $\alpha_c^{in}$ and $\alpha_c^t$ correspond to $\rho_c^{in}$ and $\rho_c^t$, respectively.

For $\hat{H}_m$, we have
\[
\hat{H}_m \rho_c^t \hat{H}_m^\dagger = \begin{pmatrix} \frac{1}{2} + \alpha_1 & (\alpha_3 - i\alpha_2)e^{2ik} \\ (\alpha_3 + i\alpha_2)e^{-2ik} & \frac{1}{2} - \alpha_1 \end{pmatrix}
\]
(14)
\[
= \begin{pmatrix} \frac{1}{2} \alpha_3 \cos 2k + \alpha_2 \sin 2k \\ -\alpha_2 \cos 2k + \alpha_3 \sin 2k \end{pmatrix}.
\]
(15)

Then we can calculate $\hat{L}^H$ by
\[
\hat{L}^H \begin{pmatrix} \alpha_1 \\ \alpha_2 \\ \alpha_3 \end{pmatrix} = \begin{pmatrix} \alpha_2 \sin 2k + \alpha_3 \cos 2k \\ -\alpha_2 \cos 2k + \alpha_3 \sin 2k \\ \alpha_1 \end{pmatrix},
\]
(16)

and obtain the expression of $\hat{L}^H$
\[
\hat{L}^H = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & \sin 2k \\ 0 & -\cos 2k & \sin 2k \end{pmatrix}.
\]
(17)

Similarly, the expression of $\hat{L}^1$ is
\[
\hat{L}^1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2k & -\sin 2k & 0 \\ 0 & \sin 2k & \cos 2k & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.
\]
(18)

3. Optimal coin sequence

The optimal coin sequence $C_T$ maps any initial coin state $\rho_c^{in} = [\theta, \phi]^{in}$ to identity state $\rho_c^t = 1/2$. In the context of superoperator, we have the following definition of optimal coin sequence.

Definition 1 (Optimal coin sequence). A coin sequence $C_T$ is the optimal sequence if its corresponding superoperators can transform $\alpha_c^{in}$ to $\alpha_c^t$, where
\[
\alpha_c^{in} = \frac{1}{2} \begin{pmatrix} 1 \\ \cos \phi \sin \theta \\ \sin \phi \sin \theta \\ \cos \theta \end{pmatrix}, \alpha_c^t = \frac{1}{2} \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}.
\]
(19)

We first propose the following optimal coin sequence with one Hadamard operation:

Theorem 1. Given $l_1, l_2 \in \mathbb{N}$, the coin sequence with $b = 1^{\otimes l_1}01^{\otimes l_2}$ is optimal if $l_1$ and $l_2$ satisfy $l_1 \neq 0$ and $l_1 \neq l_2 + 1$.

Proof. Given an $l \in \mathbb{N}^+$, $\hat{L}^l$ has property of
\[
(\hat{L}^l)^{\otimes l} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(2lk) & -\sin(2lk) & 0 \\ 0 & \sin(2lk) & \cos(2lk) & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.
\]
(20)

Then we calculate $\alpha_c^t$ by
\[
\alpha_c^t = \int_{-\pi}^{\pi} \frac{dk}{2\pi} (\hat{L}^l)^{\otimes l} \hat{L}^H (\hat{L}^l)^{\otimes l} \alpha_c^{in}
\]
(21)

where
\[
\alpha_1 = \cos \theta \cos [2(l_2 + 1)k] + \sin \phi \sin \theta \sin [2(l_2 + 1)k] \cos(2l_1k)
+ \cos \phi \sin \theta \sin [2(l_2 + 1)k] \sin(2l_1k)
\]
\[
\alpha_2 = \cos \theta \sin [2(l_2 + 1)k] - \sin \phi \sin \theta \cos [2(l_2 + 1)k] \cos(2l_1k)
- \cos \phi \sin \theta \cos [2(l_2 + 1)k] \sin(2l_1k)
\]
\[
\alpha_3 = \cos \phi \sin \theta \cos(2l_1k) - \sin \phi \sin \theta \sin(2l_1k).
\]
(22)

The momentum integrals for $\alpha_3$ is under condition $l_1 \neq 0$, and that of $\alpha_1$ and $\alpha_2$ are 0 as well under condition $l_1 \neq l_2 + 1$. Then the coin sequence with $b = 1^{\otimes l_1}01^{\otimes l_2}$ transforms $\alpha_c^{in}$ to $\alpha_c^t$ satisfying Definition 1, and is the optimal coin sequence.

We then propose another optimal coin sequence with two Hadamard operations:
Theorem 2. Given \( l_1, l_2, l_3 \in \mathbb{N} \), the coin sequence with \( b = 1 \otimes l_1 \otimes l_2 \otimes l_3 \) is the optimal coin sequence if \( l_1, l_2 \) and \( l_3 \) satisfy \( l_1 \neq l_2 + 1 \), \( l_1 \neq l_3 + 1 \), \( l_2 \pm 1 \neq l_3 \) and \( l_2 + l_3 - l_1 \neq 2 \).

Proof. \( \alpha_3^f \) is calculated by

\[
\alpha_3^f = \int_{-\pi}^{\pi} \frac{dk}{2\pi} (\hat{\mathcal{L}}_1) \otimes l_3 \hat{\mathcal{L}}_1^H \hat{\mathcal{L}}_2 \hat{\mathcal{L}}_2^H \hat{\mathcal{L}}_3 \alpha_3^\text{in} \\
= \int_{-\pi}^{\pi} \frac{dk}{2\pi} \begin{pmatrix}
1 \\
\alpha_1 \\
\alpha_2 \\
\alpha_3
\end{pmatrix}
\]

with

\[
\begin{align*}
\alpha_1 &= \cos \theta \sin [2(l_2 + 1)k] \sin [2(l_3 + 1)k] \\
- \sin \phi \sin \theta \left[ \cos(2l_1k) \cos [2(l_2 + 1)k] \sin [2(l_3 + 1)k] + \sin(2l_1k) \cos [2(l_3 + 1)k] \right] \\
- \cos \phi \sin \theta \left[ \sin(2l_1k) \cos [2(l_2 + 1)k] \sin [2(l_3 + 1)k] - \cos(2l_1k) \cos [2(l_3 + 1)k] \right]
\end{align*}
\]
\[
\begin{align*}
\alpha_2 &= - \cos \theta \sin [2(l_2 + 1)k] \cos [2(l_3 + 1)k] \\
- \sin \phi \sin \theta \left[ \cos(2l_1k) \cos [2(l_2 + 1)k] \cos [2(l_3 + 1)k] + \sin(2l_1k) \sin [2(l_3 + 1)k] \right] \\
- \cos \phi \sin \theta \left[ \sin(2l_1k) \cos [2(l_2 + 1)k] \cos [2(l_3 + 1)k] - \cos(2l_1k) \cos [2(l_3 + 1)k] \right]
\end{align*}
\]
\[
\begin{align*}
\alpha_3 &= \cos \theta \cos [2(l_2 + 1)k] + \sin \phi \sin \theta \cos(2l_1k) \sin [2(l_2 + 1)k] + \cos \phi \sin \theta \sin(2l_1k) \sin [2(l_2 + 1)k].
\end{align*}
\]

The momentum integrals for \( \alpha_3 \) is under condition \( l_1 \neq l_2+1 \), and that of \( \alpha_1 \) and \( \alpha_2 \) are 0 under condition \( l_2 \pm 1 \neq l_3 \), \( l_2 + l_3 - l_1 \neq 2 \) and \( l_1 \neq l_3 + 1 \).

The coin operation \( \hat{C}_1 \) at the first step \( (T = 1) \) is equivalent to change the initial coin state so that we have the following Corollary:

Corollary 1. Given \( l_1, l_2, l_3 \in \mathbb{N} \) satisfying Theorem 1 and Theorem 2, the coin sequences with \( b = 01 \otimes (l_1-1)101 \otimes l_2 \) and \( b = 01 \otimes (l_1-1)01 \otimes l_2 101 \otimes l_3 \) are the optimal sequence.

Experimental settings of coin sequences \( C_T \) with coin sets \( \{\hat{H}, \hat{F}\} \) and \( \{\hat{H}, \hat{F}\} \) are shown in Table I.

| \( T \) | \( C_T \) with coin set: \( \{\hat{H}, \hat{F}\} \) | \( C_T \) with coin set: \( \{\hat{H}, \hat{F}\} \) |
|---|---|---|
| 3 | \{\hat{H}, \hat{F}, \hat{H}\} | \{\hat{F}, \hat{F}, \hat{F}\} |
| 4 | \{\hat{H}, \hat{H}, \hat{F}, \hat{H}\} | \{\hat{F}, \hat{H}, \hat{H}, \hat{F}\} |
| 5 | \{\hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |
| 6 | \{\hat{H}, \hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |
| 7 | \{\hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |
| 8 | \{\hat{H}, \hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |
| 9 | \{\hat{H}, \hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |
| 10 | \{\hat{H}, \hat{H}, \hat{F}, \hat{H}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} | \{\hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}, \hat{F}\} |

Table I: Experimental settings of coin sequences \( C_T \) with coin sets \( \{\hat{H}, \hat{F}\} \) and \( \{\hat{H}, \hat{F}\} \).

Appendix B: More experimental results

The geometric representation of reconstructed \( \chi^{\exp}_{C_T} \) with coin sets \( \{\hat{H}, \hat{I}\} \) and \( \{\hat{H}, \hat{F}\} \) at \( T = 3, 5, 7 \) and 9 are shown in Fig. 5.

![Fig. 5](image-url)
Appendix C: Transport properties

One feature in QW is the trend of spreading of probabilities, which can be indicated by the second moment of the walker

\[ m(t) := \sum x^2 P(x, t). \]  
\[ (C1) \]

The walker shows a ballistic behavior if \( m(t) \propto t^2 \), while a diffusive behavior if \( m(t) \propto t \). \( m(t) \propto t^\alpha \) with \( 1 < \alpha < 2 \) indicates a supperdiffusive behavior [64]. In a 10-step QW associated with coin sequences \( C_{10} \) in Table I, we measure the probability \( P(x, t) \) at each step \( t \), and calculate the \( m(t) \) accordingly. The results of \( m(t) \) with initial coin state \( |H\rangle, |V\rangle, |+\rangle \) and \( |L\rangle \) are shown in Fig. 6, and for all three coin sets the walkers show a supperdiffusive behavior.

![Figure 6](image)

**FIG. 6.** The experimental results of \( m(t) \). The blue marks represent the results of coin set \( \{\hat{H}, \hat{I}\} \) with initial coin state of \( |H\rangle, |V\rangle, |+\rangle \) and \( |L\rangle \). The orange and yellow marks represent the results of coin \( \{\hat{H}, \hat{I}\} \) and \( \{\hat{H}\} \). Two black lines are \( m(t) = t^2 \) and \( m(t) = t \).

Appendix D: A comparison of our coin sequence with the other coin sequences.

In the context of MEG, a comparison of our coin sequence \( C_T \) with the other coin sequences including disordered coin sequence [41], Parrondo sequences [50], and three coin sequences proposed by A. Gratsea et al. [48, 49] is shown in Table II.
| Steps $^1$ | Independence $^2$ | Number of coins $^3$ | Technique $^4$ | Figure of Merit | Experimental demonstrations |
|-----------|------------------|----------------------|----------------|----------------|--------------------------|
| This work | $T \geq 3$       | Yes                  | One            | $\mathcal{F}_{C_T}$ | Linear optics (this work) |
| R. Vieira et al. (2013) [41] | $T \to \infty$ | Yes                  | Two            | Disorder       | $\langle S_E \rangle$      | Linear optics (Wang et al. (2018) [47]) |
| B. V. Govind et al. (2020) [50] | $T = 3, 5$   | Partially$^a$        | Two            | Parrondo sequences | $\langle S_E \rangle$      | None |
| A. Gratsea et al. (2020) [49] | None            | No                   | Full set of SU(2) coins | Basin hopping | $\langle S_E \rangle$ & Inverse participation ration | None |
| A. Gratsea et al. (2020) [48] | None            | Partially$^b$        | Two            | Reinforcement learning | $\langle S_E \rangle$      | None |

1 The steps that the average von Neumann entropy $\langle S_E \rangle = 1$ or the process fidelity $\mathcal{F}_{C_T} = 1$ is fulfilled.
2 The independence of initial coin state $|\theta, \phi \rangle = \cos(\theta/2)|0\rangle_c + e^{i\phi} \sin(\theta/2)|1\rangle_c$.
3 The number of coin operations in coin sequence $C_T$.
4 Techniques and algorithms to determine the coin set or coin sequence $C_T$.

$^a$ Independent of $\phi$.
$^b$ Universal sequence: independent of $\theta$ when $\phi = 0$; Optimal sequence: dependent of $\theta$ and $\phi$.

**TABLE II.** Comparison of performances of entanglement generation with different coin sequences.