Sequentially constrained Monte Carlo sampler for quantum states

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Random samples of quantum states with specific properties are useful for various applications, such as Monte Carlo integration over the state space. In the high-dimensional situations that one encounters already for a few qubits, the quantum state space has a very complicated boundary, and it is challenging to incorporate the specific properties into the sampling algorithm. In this paper, we present the Sequentially Constrained Monte Carlo (SCMC) algorithm as a powerful and versatile method for sampling quantum states in accordance with any desired properties that can be stated as inequalities. We apply the SCMC algorithm to the generation of samples of bound entangled states; for example, we obtain nearly ten thousand bound entangled two-qutrit states in a few minutes—a colossal speed-up over independence sampling, which yields less than ten such states per day. In the second application, we draw samples of high-dimensional quantum states from a narrowly peaked target distribution and observe that SCMC sampling remains efficient as the dimension grows. In yet another application, the SCMC algorithm produces uniformly distributed quantum states in regions bounded by values of the problem-specific target distribution; such samples are needed when estimating parameters from the probabilistic data acquired in quantum experiments.

I. INTRODUCTION

Random samples of quantum states with specific probability distributions and/or governed by specific physical properties are very useful for many applications in quantum information science and other research areas. For instance, they play an important role in quantum state and parameter estimation [1–3], allowing one to test and verify properties for certain classes of quantum states [4–9], as well as to test quantum channels and processes, such as the random quantum circuits leading to Quantum Supremacy [7–9]. Similar to sampling classical systems, independent rejection sampling methods are often inapplicable to high-dimensional quantum systems as they severely suffer from the exponential growth of resource requirements, known as the “curse of dimensionality” [10]. The positivity boundaries of the quantum state spaces, moreover, are typically extremely complex to characterize for otherwise convenient parameterizations, causing quantum states to be more challenging to sample.

Most existing methods for the direct sampling of the random density matrices, which represent the quantum states numerically, rely on either clever parameterization of the state space or various kinds of induced measures [11–15]. The major drawback is that the exact probability distribution for random states constructed this way are usually not known; one has to estimate approximate distributions, and doing this reliably is a numerical challenge in itself. Thus, rejection sampling methods that proceed from known proposal distributions are still often favored for sampling from a desired target distribution. However, even if one can directly draw from a reliable reference distribution that well mimics the target distribution—for example, using a version of the complex Wishart distribution [16–18]—the exponential reduction of the acceptance rate as the dimension increases makes it rather impractical to go beyond sampling small quantum systems (see, for example, Ref. [19]).

Monte-Carlo-based algorithms [20] can be much more resilient against this curse of dimensionality. The major difficulty encountered in using Monte Carlo methods comes from the incorporation of constraints into the parameter space. For instance, enforcing the positivity constraint needed for a physical state, which is intrinsic to quantum systems, can be extremely challenging. Algorithms that rely on clever random walks in the probability simplex, such as the Markov Chain Monte Carlo (MCMC), suffer from low acceptance rates because of this positivity constraint, and the Hamiltonian Monte Carlo (HMC) algorithm [21], whose random walk stays in the state space, depends on the evaluation of a Jacobian determinant and its derivatives, found to be numerically unreliable in a high-dimensional quantum state space [22–23].
In this work, aiming at overcoming the curse of dimensionality as well as at efficiently enforcing quantum constraints, we propose the use of the sequentially constrained Monte Carlo (SCMC) samplers for the sampling of quantum states in accordance with a given distribution and/or certain physical properties. The SCMC samplers were first proposed by Golchi and Campbell in 2016 [24] to effectively impose constraints when sampling classical systems as an extension to the sequential Monte Carlo (SMC) samplers proposed by Del Moral et al. a decade earlier [25]. SMC methods [26–28], which have been used extensively in the context of sequential Bayesian inference, are not new to the field of quantum information. For example, they were well used in adaptive/online Bayesian Hamiltonian estimation [29–31, 32], and so confirm that the direct SCMC samplings from the target distribution is reliable. In addition, we observe that the direct SCMC sampling is more efficient than the indirect OTJ method. Depending on one’s point of view, the last example can be regarded as benchmarking SCMC against OTJ or OTJ against SCMC.

Samples and codes can be fetched from a dedicated repository [33].

II. SCMC SAMPLING

The SMC samplers presented in Ref. [25] enable one to sample \( \mathbf{x} \) efficiently from its difficult-to-sample target distribution \( f(\mathbf{x}) \) through a sequence of intermediate distributions. We choose an initial distribution \( g(\mathbf{x}) \), which can be the prior or any appropriate reference distribution that is easy to sample from directly, and find a discrete sequence of \( N_\tau \) density distributions \( \{g_i(\mathbf{x})\}_{i=0}^{N_\tau} \) that smoothly bridges between \( g(\mathbf{x}) \) and \( f(\mathbf{x}) \).

One particular choice of \( g_i(\mathbf{x}) \) is to follow the geometric path [34, 35],

\[
h_i(\mathbf{x}) = f(\mathbf{x})^{\tau_i} g(\mathbf{x})^{(1-\tau_i)},
\]

where \( \tau_i \) runs from 0 to 1 in arithmetic progression, i.e., \( \tau_i = i/N_\tau \) as \( i \) goes from 0 to \( N_\tau \). Thus, \( h_0(\mathbf{x}) = g(\mathbf{x}) \) and \( h_{N_\tau}(\mathbf{x}) = f(\mathbf{x}) \).

The utility of SMC samplers was extended by Golchi and Campbell [24] to SCMC samplers; in their approach the hard constraints on the parameter space are gradually incorporated into the intermediate distributions as soft probabilistic constraints. Here, soft constraints refer to relaxations that approach the hard constraint in the limit. For instance, when the desired hard constraint is \( \kappa(\mathbf{x}) > 0 \) with its indicator function being the step function \( I_\kappa(\mathbf{x}) = \eta(\kappa(\mathbf{x})) \), the intermediate indicator functions for the soft constraints can be smooth approximations of the Heaviside step function, such as

\[
I_{\kappa,i}(\mathbf{x}) = \frac{1 + \tanh(a \tau_i \kappa(\mathbf{x}))}{2},
\]

where the tolerance \( a \) is an adjustable scale controlling the hardness of the constraint. In this case, the acceptance rate in the MCMC iterations is directly related to the value of the indicator function \( I_{\kappa,i}(\mathbf{x}) \) by taking

\[
h_i(\mathbf{x}) \rightarrow h_i(\mathbf{x}) I_{\kappa,i}(\mathbf{x}),
\]

and, in the limit of \( a \tau_i \rightarrow \infty \), it converges to the hard constraint, i.e.,

\[
I_{\kappa,N_\tau}(\mathbf{x}) \rightarrow \begin{cases} 
1 & \text{for } \kappa(\mathbf{x}) > 0, \\
0 & \text{for } \kappa(\mathbf{x}) < 0.
\end{cases}
\]

In practice, for a large but yet finite value of \( a \), points with \( \kappa(\mathbf{x}) \leq 0 \) (\( \kappa(\mathbf{x}) > 0 \)) are rejected (accepted) with a higher and higher probability as \( \tau_i \rightarrow 1 \), thus sample points gradually move towards the region satisfying the constraint. At the final step of the SCMC algorithm, we impose the hard constraint instead of the probabilistic constraint and reject the sample points that violate the constraint. Consult the Appendix for implementation details.
The ability to enforce constraints via SCMC is very useful for sampling quantum states, because the quantum Hilbert space intrinsically requires the physicality constraint which usually means complicated constraints on the parameters (positivity of a large matrix) that are CPU-expensive to check. Apart from the physicality constraint, many other interesting quantum constraints can also be extremely difficult to enforce as suitable parameterizations of the states are unavailable. For efficient sampling of quantum states, we need to identify the quantum constraints and find their corresponding indicator functions. Next, we show how well the SCMC sampler works for sampling quantum states in three distinct contexts.

III. EXAMPLES

A. Bound entanglement

A bound entangled state is a state with non-distillable entanglement. It has drawn a lot of attention in the quantum information community since the prediction of its existence in 1998 [36]. Random samples of bound entangled states and the study of them is useful not only for the field of quantum information but also for mathematical interests. On the one hand, states with bound entanglement provide a testbed for studying the relation between entanglement, steering, and Bell-type nonlocality [37, 38]; on the other hand, bound entangled states enable the study of positive maps from a different perspective [39]. Moreover, despite being highly mixed, such states are potentially useful for quantum cryptography and quantum metrology [40, 41].

While it is currently unknown if all bound entangled states have a positive partial transpose (PPT), we do know that all entangled PPT states are bound entangled [36], and a PPT state is surely entangled if the cross norm or realignment (CCNR) criterion is met [42]. Taken together, then, the two conditions

$$\min \{\mu_{PT}\} \geq 0 \quad \text{and} \quad \sum_{j} \sigma_j(\tilde{\rho}) = R > 1 \quad (5)$$

are sufficient to ensure bound entanglement. Here, $\{\mu_{PT}\}$ is the set of eigenvalues of the partial transpose of the density matrix $\rho$ and $R$ is the sum of the singular values of the realigned matrix $\tilde{\rho}$ [42, Eq. (5)]. Examples of bound entangled states are studied in the literature; see, e.g., Refs. [36, 37, 43, 49]. They are often given as special constructions of states in some particular parameter families. A more general way of constructing bound entangled states which allows the generation of a random sample has also been presented in Ref. [50], but it only works for bipartite systems with equal dimensions.

The SCMC samplers are well suited for generating random samples of PPT bound entangled states with no restrictions on the states’ parameter family or on their dimensionality. They can be generated by imposing the two criteria in Eq. (5) in the same way as one imposes the physicality constraint. The two indicator functions are in the form of Eq. (2) with $\kappa_1(\rho) = \min \{\mu_{PT}\}$ and $\kappa_2(\rho) = R - 1$, respectively. The initial reference sample of dimension $d$ can be drawn directly from a uniform distribution of physical states using the Wishart distribution $W_{d+2}(d, 1_d)$, that is, they are generated from $d \times d$ normally distributed complex matrices with mean zero and covariance matrix $1_d \otimes 1_d$ (see Ref. [19] for more details). Since the reference samples are physical initially, and we do not want to have unphysical sample points as the samples are processed, we impose the hard physicality constraints during the MC steps to ensure that the random walks do not move a quantum state beyond the physical boundary. Examples illustrating the generation of random samples of bound entangled states are shown in Fig. 1 where the states in the initial reference sample are represented by red crosses and the sample states after SCMC and before the final rejection sampling are represented by blue dots. As is clearly visible from the plots, our SCMC sampler successfully moved the states toward the region around $R = 1$ and $\min \{\mu_{PT}\} = 0$ and produced bound entangled states.

As discussed in the previous section, the efficiency of SCMC depends on a number of parameters, including the tolerance $a_\rho$ for the PPT constraint, the tolerance $a_\tau$ for the CCNR entanglement constraint, and the number $N_T$ of the distribution steps. Rejection sampling is applied to impose the complete set of hard constraints at the end of the algorithm, thus a good set of the parameters should result in a high yield of accepted sample points within reasonable CPU time. For the bipartite qutrit system of dimension $d = 3 \times 3$, a 99% yield of bound entangled states was obtained by setting $N_T = 300$ and $a_\tau = a_\rho = 5 \times 10^4$. On our standard desktop, it took a few minutes to find thousands of two-qutrit bound entangled states; CPU parallelization is possible and will speed up the computation. For comparison, we conducted independent sampling by drawing $10^{10}$ states from a uniform distribution of bipartite qutrit states. Only 24 out of the $10^{10}$ states obeyed the criteria of Eq. (5), and this process took days even with CPU parallelization. Such a search for higher-dimensional systems ($3 \times 4, 3 \times 5, \text{ or } 4 \times 4, \text{ say}$) is even more difficult. This evidently shows that the improvement provided by the SCMC algorithm is tremendous.

In Fig. 1 we show scatter plots of the $\min \{\mu_{PT}\}$ and $R$ values of samples obtained by SCMC for five systems with Hilbert-space dimensions between eight and sixteen (state space dimensions between 63 and 255). No exhaustive trials were done, and the performance can certainly be improved upon as we did not optimize the parameters. Since we are imposing the constraints through the two inequalities in Eq. (5), the states produced were clustered in the small corner near $R = 1$ and $\min \{\mu_{PT}\} = 0$, except for the $2 \times 4$ system. To explore other parts of the space for larger values of $R$ and $\min \{\mu_{PT}\}$, we filtered the
FIG. 1. The generation of bound entangled states using SCMC. The initial reference sample has $10^4$ states (red crosses) uniformly distributed with respect to the Hilbert–Schmidt distance shown here on an $R$ vs. min$\{\mu_{PT}\}$ plot. The states after SCMC are indicated by the blue dots. Out of these 10,000 post-SCMC states, 8,530, 7,011, 2,211, and 4,013 states are bound entangled for the respective systems of dimensions $3 \times 3$, $3 \times 4$, $3 \times 5$, and $4 \times 4$ (top and middle rows). For these, the insets show samples filtered through further MCMC iterations with different propagation kernels (by choosing different directions of the random walks) represented by dots of different colors. The case of the $2 \times 4$ system (bottom left) is different: There are no blue dots in the first quadrant (gray) where the criteria of Eq. $5$ are obeyed. Enforcing only the PPT criterion results in the orange sample; enforcing only the CCNR criterion results in the green sample. The inset shows the orange sample then CCNR, first PPT, and, in different colors, further samples obtained by SCMC steps toward enforcing the CCNR criterion. The table reports the parameters used for the SCMC sampling, the yield of the procedure (i.e., the fraction of initial sample states that are turned into states in the first quadrant), and the CPU time consumed by one distribution step.

| system $| N_r | a_e | a_p | yield | time |
| --- | --- | --- | --- | ---- | ---- |
| $3 \times 3$ | 20 | 1,000 | 10,000 | 85% | 11s |
| $3 \times 4$ | 2,000 | 1,000 | 10,000 | 70% | 14s |
| $3 \times 5$ | 2,000 | 3,000 | 20,000 | 22% | 15s |
| $4 \times 4$ | 2,000 | 3,000 | 500,000 | 40% | 22s |
| $2 \times 4$ | 20 | 200 | 300 | 0% | 7s |
sample further through MCMC iterations with different kernels and acceptance criteria; the samples obtained are shown in the insets of Fig. 1. This way of producing random samples of bound entangled states, without relying on special constructions of parameter families of states, will be very useful when studying general properties of bound entanglement; such investigations are, however, not the objective of this work.

The $2 \times 4$ system is particular—we could not find any states that satisfy both the PPT and the CCNR criterion in Eq. (5). When either one of the criteria is enforced, SCMC yields samples with $\min \{\mu_{PT}\} \geq 0$ or $R > 1$, respectively. As the $2 \times 4$ plot in Fig. 1 shows, first enforcing the PPT criterion, followed by SCMC steps toward enforcing the CCNR criterion, produces samples with $R$ values that are sequentially closer to the $R = 1$ threshold without, however, crossing it. We conjecture that $2 \times 4$ systems do not have bound entangled states that obey the sufficient double-criterion of Eq. (5). Indeed, the families of bound entangled $2 \times 4$ states constructed by the authors of [43, 46, 47] are PPT with $R \leq 1$; their values are on the straight lines with endpoints $(\min \{\mu_{PT}\}, R) = (0, 0)$ and $(0, 0.7866), (0.0270, 0.7572)$, or $(0.1)$ and $(0.8536)$, respectively.

These examples illustrate that the criteria of Eq. (5) are not necessary—the pair is sufficient. It is possible, perhaps likely, that the final SCMC samples (blue dots in Fig. 1) contain bound entangled states outside the first quadrant, but the yields reported in the table refer solely to the states that meet the double criterion.

### B. Desired target distribution

Random samples from a desired distribution are useful in various contexts in quantum information science such as studying properties of a quantum system, parameter optimization, or model testing. Owing to the notorious curse of dimensionality and/or quantum constraints, sampling methods that work well for systems of low dimension fail to work in practice as the dimension gets larger. (The curse refers to the dimensionality of the parameter space, not of the Hilbert space.) For example, the generation of a target sample from a uniform reference distribution of states through rejection sampling fails to work for three-qubit systems as the acceptance rate is extremely low; one can increase the acceptance rate by orders of magnitude by replacing the uniform reference sample with an appropriate Wishart distribution of states, nevertheless, the efficiency still suffers from an exponential decay with respect to dimensionality [19]. On the other hand, MC algorithms suffer from instability and low efficiency issues as the dimension of the quantum system gets larger, in addition to their unavoidable sample correlation [22, 23]. In this section, we demonstrate that SCMC can be used for generating samples of a desired target distribution in high dimensions, impractical otherwise, and this algorithm does not suffer from the curse of dimensionality.

To be specific, we consider the generic situation where the desired distribution is of the Dirichlet form

$$f(\rho) \propto \prod_k \text{tr}(\Pi_k \rho^\alpha_k) = \prod_k \rho_k^\alpha_k, \quad \rho > 0,$$

(6)

which is central to quantum state estimation with a conjugate prior. $\{\Pi_k\}$ is an informationally complete probability-operator measurement (POM) with $\Pi_k \geq 0$ and $\sum_k \Pi_k = 1$, giving a one-to-one correspondence between the state $\rho$ and the probabilities $\{p_k\}$. Note that a sample drawn from the Dirichlet distribution by one of the standard efficient algorithms, which draw from the probability simplex, has many unphysical entries $(p_k > 0$ while $\rho \not\geq 0)$ and simply discarding them would result in a very poor yield. The SCMC procedure pushes most of the initially unphysical sample points into the physical state space. Similar remarks apply to samples drawn from a Dirichlet distribution centered at the state for which $f(\rho)$ is largest, often a rank-deficient state. Note also that the Dirichlet distribution has a single very narrow peak when $A = \sum_k \alpha_k$ is large, as is the typical situation in quantum state estimation scenarios.

To apply SCMC, we first need a set of initial sample points that can be easily generated. Conventional MC methods often use samples generated in the probability simplex, such as the Dirichlet distribution that do not respect the positivity constraints, but the rate of physical samples decreases exponentially with respect to the dimension of the parameter space which makes it impractical for systems of high dimension (for example, the physical rate is less than $10^{-18}$ for a three-qubit system [19]). Therefore, for efficient sampling, it is expedient to use an initial reference sample that is physical. We mainly explore two types of initial reference sample distributions — the uniform distribution with respect to the Hilbert–Schmidt distance and the peaked Wishart distribution. The former rarely resembles any feature of the target distribution but the intermediate distributions $\{h_i(\rho)\}$ are straightforward to evaluate for each sample point in the algorithm. The Wishart distribution for quantum states $W_d(n, \Sigma)$ offers more freedom by adjusting the covariance matrix $\Sigma$ in shaping the reference distribution towards a target while conforming to the physicality constraint. Its probability density for a $d$-dimensional system is [19]

$$g(\rho) \propto \frac{\det(\rho)^{n-d} \text{tr}(\Sigma^{-1} \rho)^{k-d}}{\text{tr}^d(\Sigma^{-1} \rho)},$$

(7)

where $\rho = Z^\dagger Z / \text{tr}(Z^\dagger Z)$ and the $d \times n$ random complex matrix $Z$, with $n \geq d$, is drawn from a gaussian distribution with zero mean and covariance matrix $1_n \otimes \Sigma$; we get the uniform distribution for $n = d$ and $\Sigma = 1_d$. We expect faster convergence using the Wishart distribution as it is more similar to the target distribution. However, the apparent downside is that the numerical evaluation of
its initial and resulting intermediate distributions takes longer.

To test its performance, we run the SCMC algorithm for different numbers of intermediate distributions $N_\tau$ and different types of initial reference distributions. We assess the quality of the sample by a variant of the method described in [19], Sec. 5, for which we introduce the nested $\lambda$-regions with $f(\rho) \geq \lambda F$ where $F = \max_{\rho} \{ f(\rho) \}$ is the peak value of $f(\rho)$, and $0 \leq \lambda \leq 1$. We have the full physical state space for $\lambda = 0$ and only the peak location for $\lambda = 1$. The fraction of the target distribution contained in a $\lambda$-region is its content $c_\lambda$:

$$c_\lambda = \int \left( \int \rho d\rho \right) \eta(f(\rho) - \lambda F),$$

where $c_{\lambda=1} = 1$ reflects the normalization of $f(\rho)$ to unit integral. By counting how many sample points are inside a $\lambda$-region, we obtain an estimate for the respective $c_\lambda$ value.

We expect that this estimate is better (i) when more intermediate step $N_\tau$ are used in the SCMC algorithm, and (ii) when the sample size $N_s$ is larger. Both expectations are confirmed by the data presented in Fig. 2 for two examples, one for a three-qubit system and one for a four-qubit system. The two $f(\rho)$s are posterior distributions (for a uniform prior) for $A = 3000$ randomly generated measurement clicks of product tetrahedron POMs.

With this large $A$ value, the distributions are squeezed into a tiny region in the immediate vicinity of the peak location of $f(\rho)$ at or near the boundary of the state space, which makes them impractical for sampling with conventional methods. In the SCMC algorithm, besides imposing the sequence of intermediate distributions to approach the desired one gradually from the reference distribution, the physicality constraints also have to be imposed either strictly or gradually. When the initial sample points are guaranteed to be physical, such as the ones uniformly drawn from the state space, the hard physicality constraint is directly imposed during the MCMC iterations. Otherwise, when a portion of the initial sample distribution is unphysical, as in the case of the linearly shifted Wishart distribution or the Dirichlet distributions, we impose the soft physicality constraint gradually along with the distribution steps.

In the three-qubit example shown in Fig. 2(a), we find it sufficient to have $N_\tau = 300$ intermediate steps when the initial sample is drawn from an appropriate Wishart distribution, as the content barely changes by increasing $N_\tau$ further. When $N_\tau = 100$, the Dirichlet distribution centered at the peak of $f(\rho)$ performs the best and the uniform distribution performs the poorest. Their difference becomes less and less noticeable with increasing number of distribution steps. For example, no significant difference of their performance is shown when $N_\tau = 150$. Thus, it is evident that the SCMC algorithm tolerates flexibility in the choice of the initial reference distribution, although a reference distribution that better resembles the target can offer faster convergence.

Figure 2(b) shows the content of the four-qubit samples generated starting from either a Wishart distribution or a uniform distribution. The Dirichlet distribution is impractical to use here because the random walk into the physical space—a tiny subspace within the 255-dimensional probability simplex—is extremely inefficient. For the same number of sample points, the Wishart distribution might appear to provide faster convergence than the uniform distribution in terms of $N_\tau$. However, the overall performance using the uniform distribution is practically better. This is because the computation time
per sample point is about three to four times faster using the uniform distribution than for the Wishart distribution. A larger sample size helps not only to reduce the statistical error in evaluating sample average quantities like the content, it also makes the random walk, which is set to propagate along the direction given by the covariance matrix of the current sample, more efficient [54]. As a result, the time cost for using the Wishart distribution with \( \{N_s = 3 \times 10^4, N_r = 3500\} \) is about the same as using the uniform distribution with \( \{N_s = 10^5, N_r = 3500\} \) but the latter converges better, as is visible in Fig. 2(b).

The computational time roughly scales as \( O(N_r \sqrt{N_s}) \) with simple vectorization of the code in Python, provided that there is sufficient memory. For the examples shown in Fig. 2 the sampling of \( 10^5 \) three-qubit states and \( N_r = 200 \) distribution steps took about \( 2 \times 10^4 \) seconds using the Wishart distribution on a regular desktop with no CPU-parallelization, and \( 7 \times 10^3 \) seconds using the uniform distribution. The sampling of \( 10^5 \) four-qubit states and \( N_r = 3500 \) distribution steps using the uniform distribution, which showed good convergence, took about \( 2.2 \times 10^5 \) seconds (\( \sim 60 \) hours, which is only about 30 times longer than sampling three-qubit states). We also ran the sampling algorithm for one- and two-qubit states with reliable sample verification. Using the uniform distribution, the sampling of \( 10^5 \) one-qubit states and \( N_r = 10 \) takes about 260 seconds and the sampling of \( 10^5 \) two-qubit states and \( N_r = 75 \) takes about \( 1.6 \times 10^3 \) seconds [51]. Similar scaling of running time was seen in other distributions we sampled. All sampling is done on a standard desktop with 8 GB RAM.

In summary, for the SCMC sampling from the states spaces for one to four qubits, which have \((3, 15, 63, 255)\) parameters, the number of required distribution steps \( N_r \) is \((10, 75, 200, 3500) \approx (3^{2.1}, 15^{1.6}, 63^{1.3}, 255^{1.5}) \) and the computational time is roughly \((3^{1.3}, 15^{1.2}, 63^{1.2}, 255^{1.5}) \) minutes. The number of distribution steps required for convergence increases as the dimensionality increases, leading to longer computational time, and additional CPU cost is incurred by the multiplication of ever larger matrices. For the scaling of the running time, we observe exponents that are approximately independent of the dimension, and this suggests that the SCMC algorithm does not suffer an exponential increase in computation time with respect to the dimension as other sampling methods do. Put differently, the SCMC sampler appears to remain computationally efficient as the dimensionality increases, at least for dimensions up to 255. Owing to limited time and computer memory, we did not sample larger quantum systems. However, a powerful work station running the CPU-parallelization version of the code should be able to not only produce a larger sample in a much shorter period of time but also sample systems of higher dimensions. There is huge potential in the SCMC algorithm which is worth exploring.

C. The Oh–Teo–Jeong method for benchmarking

The volume element \((d\rho)\) in Eq. 8 is also the probability element of the uniform distribution, and

\[
s_\lambda = \int (d\rho) \eta(f(\rho) - \lambda F)
\]

is the size of the \( \lambda \)-region, normalized such that \( s_{\lambda=0} = 1 \). The relation [52]

\[
c_\lambda = \frac{\lambda s_\lambda + \int_0^1 d\lambda' s_{\lambda'}}{\int_0^1 d\lambda' s_{\lambda'}}
\]

can be used to obtain an estimate of \( c_\lambda \) from an estimate of \( s_\lambda \). Owing to the very narrow peak of \( f(\rho) \), however, we cannot estimate \( s_\lambda \) by counting how many points of a uniform sample are in the \( \lambda \)-region, there would be far too few sample points in the vicinity of the peak.

The OTJ method introduced in [31, 32] offers a clever solution to this problem based on their region-average computation lemma. It provides \( s_\lambda \) by an integration,

\[
s_\lambda = s_{\lambda_0} \frac{g_{\lambda_0}}{g_\lambda} \exp \left(- \int_{\lambda_0}^{\lambda} \frac{d\lambda'}{\lambda g_{\lambda'}} \right),
\]

where \( g_\lambda \) is the average value of \( \ln(f(\rho)/(\lambda F)) \) over the \( \lambda \)-region with respect to the uniform distribution. Although \( f(\rho) \) is very narrowly peaked, \( \ln(f(\rho)) \) is not, and \( g_\lambda \) can be estimated accurately by a sufficiently large sample drawn from the uniform distribution over the \( \lambda \)-region, not over the whole quantum state space. Accordingly, an implementation of the OTJ method requires an accurately known reference value \( s_{\lambda_0} \) and a reliable procedure for generating a uniform sample for each \( \lambda \)-region. Both ingredients had problems in the implementation reported in [31, 32].

Here we implement the OTJ method by (i) finding \( s_{\lambda_0} \) from a large uniform sample for \( \lambda_0 \) small enough that a few percent of the sample points are in the \( \lambda_0 \)-region; and (ii) generating large uniform samples from successive \( \lambda \)-regions by an SCMC algorithm that gradually imposes the constraint \( f(\rho) \geq \lambda F \). For the computation of the \( \lambda' \) integral in Eq. 11 we discretize linearly in \( \ln(\lambda') \).

Figure 3 illustrates aspects of the OTJ algorithm. On the left we see that the \( \lambda \)-regions are not of ellipsoidal shape [57] and that one needs truly tiny positive \( \lambda \) values to enclose a sizable fraction of a uniform sample on the whole state space. The situation is that of a three-outcome measurement in which \( A = 2420 \) events were observed [58, p.224]. While this is not a lot of data for a one-qubit measurement, the Bayesian posterior (our target function here) is already peaked extremely narrowly.

On the right in Fig. 3 we show \( c_\lambda \) for a three-qubit target distribution obtained from sampling directly from the distribution by SCMC (black curve) and by different runs of the OTJ algorithm (colored curves). Figure 3(right)
confirms that the SCMC sampling yields the correct $c_\lambda$ values, as the sequence of OTJ curves converges toward the SCMC curve. This convergence is from below, which tells us that the OTJ estimates of $c_\lambda$ have a negative bias.

All SCMC runs of the OTJ method proceed from $\lambda = 1$ when all sample points are at the peak location. Then by reducing $\lambda$ in $N_\lambda$ steps, we obtain uniform samples on the respective $\lambda$-regions. The “calibration runs” then use $s_{\lambda_0} = 0.024$ for $\lambda_0 = 10^{-250}$, which we get from a large uniform sample on the whole state space, in Eq. (11) and the resulting $s_\lambda$ in Eq. (10). The “precision runs” use $\lambda_1 = 10^{-25}$ in the stead of $\lambda_0$ in Eq. (11), with $s_{\lambda_0} \simeq 10^{-24}$ obtained from the calibration runs, and $N_\lambda'$ intermediate $\lambda$ values. The graphs for the final calibration run and the first precision run, marked by * in Fig. 3, are indistinguishable.

Regarding the computational effort, the comparison of the SCMC and the OTJ algorithms speaks strongly in favor of SCMC. The computation time scales roughly as $\mathcal{O}((N_\lambda + N_\lambda')N_\tau \sqrt{N_\tau})$. Therefore, the final precision run takes about $(2000+6000)50\sqrt{10^{6}}/(200\sqrt{10^{8}}) \simeq 10^{2}$ times longer than the direct production of a target sample by SCMC to evaluate $c_\lambda$, and the latter has higher accuracy.

In summary, we can produce samples uniformly distributed in $\lambda$-regions by a SCMC implementation of the OTJ method, and so evaluate the content $c_\lambda$ as a function of $\lambda$. Our numerical results not only confirm that the OTJ method is viable, it also demonstrates that, for a sharply peaked target distribution, it is more efficient to evaluate the content from the target sample produced directly by SCMC than using the more involved OTJ method. Further, we observe that the content estimate obtained by the OTJ method has a negative bias.

IV. CONCLUSION

In conclusion, we demonstrate the reliability and efficiency of the SCMC sampler in sampling quantum states through three explicit examples. In the first example, we produce samples of bound entangled states for dimensions $3 \times 3$, $3 \times 4$, $3 \times 5$, and $4 \times 4$. For some of these cases, no such samples were available before, and our method can also provide bound entangled states for higher dimensions. For example, we obtain nearly ten thousand two-qutrit bound entangled states in a few minutes by SCMC sampling, whereas several days of independence sampling yield 24 such states from $10^{10}$ candidates. We also observe that the $2 \times 4$ system is particular, as we could not find bound entangled states that obey the PPT-CCNR double criterion. Based on the numerical evidence, we conjecture that such states do not exist.

In the second example, we use the SCMC method for sampling quantum states in accordance with a given target distribution three-qubit and four-qubit systems. We find that this algorithm is more efficient than others, and it appears to remain efficient even as the dimension grows.

The third example is a SCMC implementation of the OTJ method for producing uniform samples on regions bounded by values of the target function (the $\lambda$-regions); the shortcomings of the original implementation are avoided. We find that, for the purpose of estimating the content of the $\lambda$-regions, direct SCMC sampling from the target distribution is much more efficient, and the values obtained by the OTJ method have a negative bias.

The SCMC sampler can be applied to many other sampling problems of quantum systems as long as the con-
straints can be described by inequalities. For example, one could produce samples of states that violate an inequality of the Bell kind. Multiple constraints can be efficiently applied in parallel. Moreover, due to the well-known channel-state duality, SCMC sampling can also be used to sample channels. We invite the readers to apply the method to their specific sampling problems.

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APPENDIX: SCMC ALGORITHM

The general algorithm takes in a set of $N_s$ initial sample points with distribution $g(x)$ and finds a discretized sequence of density distributions $\{h_i(x)\}$ that “smoothly” bridges between $g(x)$ and $f(x)$. $\{h_i(x)\}$ takes into account both the change of the probability density distribution and the incorporation of the soft constraints given by Eqs. (1)–(3).

In general, there can be $N_{MC}$ number of MCMC iterations between each distribution step and importance resampling is performed whenever the effective sample size (ESS) drops below a chosen threshold $N_{\text{thresh}}$. We employ the iteration kernel in the Metropolis algorithm which results in sample points with updated weights for the $k$th iteration $w_i^{(k)} \propto [h_i(x_i^{(k)})]/[h_i-1(x_i^{(k)})]$. The ESS, that is $\text{ESS}^{-1} = \sum_{k=1}^{N_s} (w_i^{(k)})^2$, is a measure of sample degeneracy [59], i.e., a small ESS indicates only a small portion of the sample points are filtered into the next step. This degeneracy can be much suppressed in SMC with finer distribution steps as compared with MCMC. In addition, the SMC algorithms can be conveniently implemented with parallel computation for speed-up. The general outline of our SCMC algorithm is as follows:

Sequentially constrained Monte Carlo algorithm

1: Generate an initial sample $\{\rho^{(1:N_s)}\}$ with distribution $g(\rho)$
2: Assign weight $w_0^{(k)} = 1/N_s$ for $k = 1 : N_s$
3: for $i = 1 : N_r$ do
4: Evaluate $h_i(\rho^{(1:N_s)})$ with indicators $I_{\kappa,i}(\rho^{(1:N_s)})$
5: for $j = 1 : N_{MC}$ do
6: if $j = 1$ then
7: Update $w_i^{(k)} = w_{i-1}^{(k)} \frac{h_i(\rho^{(k)})}{h_{i-1}(\rho^{(k)})}$ for $k = 1 : N_s$
8: Normalize $w_i^{(k)}$
9: Calculate ESS $= \sum_{k=1}^{N_s} (w_i^{(k)})^{-2}$
10: if ESS < $N_{\text{thresh}}$ then
11: Importance resampling according to $w_i^{(1:N_s)}$
12: Reset $w_i^{(1:N_s)} = 1/N_s$
13: end if
14: end if
15: Propagate $\{\rho^{(1:N_s)}\}$ with MCMC transition kernels
16: Accept/reject the new points
17: end for
18: end for

Figure 4(left) illustrates a simple one-dimensional example of SMC. The target distribution $f(x)$ has two peaks and we sample it by starting with random samples drawn from a single-peak normal distribution $g(x)$. A simple example that illustrates how the physicality constraint can be effectively imposed is shown in Fig. 4(right). The reference sample is uniform in the probability simplex of the distorted trine measurement.

FIG. 4. Simple illustrations of SCMC in 1D and 2D. Left: A single-peak reference sample is used to sample a target distribution with two peaks. The intermediate distributions are given by Eq. (1) with $N_r = 10$. Both the exact and the numerically estimated distribution for $10^3$ sample points are shown. Right: The physicality constraint is gradually imposed through $N_r = 10$ intermediate distribution steps. The triangle represents the boundary of the probability simplex, while the circle represents the boundary of the physical region.
of Ref. [55] on a single-qubit system and it is filtered through $N_r = 10$ intermediate constrained uniform distributions thereby imposing the physicality constraint by taking $\kappa(\rho)$ to be the smallest eigenvalue of the density matrix of state $\rho$.

The computational efficiency of the SCMC algorithms depends on a set of parameters—the number of reference sample points $N_s$, the number of distribution steps $N_r$, the number of MC iterations $N_{\text{MC}}$ that the entire sample is advanced by a kernel of choice in each distribution step, and the scaling parameter $a$—which need to be calibrated for the sampling problem at hand. For most of the examples presented in this paper, we fix $N_{\text{MC}} = 15$ [60] and adjust other parameters in accordance with the criterion set out in Ref. [61]. To reduce the sample degeneracy without compromising its efficiency by too much, we set the threshold of ESS at $N_{\text{thres}} = \frac{4}{5}N_s$ and importance resampling is executed once ESS falls below $N_{\text{thres}}$. The sample correlation resulting from importance resampling is executed once ESS falls below $N_{\text{thres}}$. To reduce the correlation from the final importance resampling before the SCMC algorithm ends, we filter the system through 20 additional MC iterations.

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The sequence of randomly generated measurement clicks data ($A = 3000$) for product tetrahedron POMs in lexicographic order: the data for the one-qubit example are $\{1135, 1086, 394, 385\}$; the data for the two-qubit example are $\{215, 34, 229, 248, 52, 244, 235, 256, 257, 225, 234, 41, 217, 238, 44, 231\}$; the data for the three-qubit example are $\{36, 13, 64, 71, 14, 16, 7, 15, 60, 10, 84, 63, 64, 9, 55, 71, 8, 12, 10, 16, 16, 48, 67, 62, 9, 64, 75, 63, 10, 74, 60, 73, 65, 14, 62, 66, 9, 57, 76, 53, 82, 78, 128, 22, 61, 44, 25, 27, 56, 12, 52, 66, 14, 76, 56, 78, 45, 47, 22, 27, 66, 68, 25, 102\}$; and the data for the four-qubit example are $\{5, 5, 11, 9, 5, 3, 5, 11, 4, 18, 18, 11, 3, 10, 24, 5, 4, 2, 5, 1, 5, 2, 2, 1, 3, 1, 5, 6, 3, 4, 16, 3, 24, 15, 3, 4, 0, 16, 4, 27, 14, 13, 1, 17, 20, 9, 4, 16, 16, 4, 2, 2, 4, 21, 3, 27, 19, 14, 18, 35, 32, 3, 1, 6, 0, 4, 0, 4, 5, 4, 6, 7, 4, 3, 3, 1, 6, 0, 4, 4, 4, 0, 15, 14, 5, 2, 5, 19, 17, 3, 8, 14, 12, 2, 2, 2, 1, 16, 13, 3, 26, 26, 21, 7, 27, 13, 13, 2, 1, 2, 4, 2, 8, 17, 21, 2, 23, 21, 13, 4, 24, 21, 18, 7, 5, 17, 19, 4, 1, 9, 1, 10, 3, 25, 17, 19, 4, 15, 18, 1, 6, 1, 1, 3, 15, 15, 17, 3, 20, 26, 21, 1, 15, 21, 17, 14, 5, 41, 16, 10, 14, 40, 23, 35, 22, 71, 20, 14, 22, 17, 3, 13, 0, 16, 17, 1, 9, 12, 23, 16, 17, 20, 1, 14, 27, 2, 17, 12, 3, 17, 20, 4, 6, 4, 3, 17, 4, 19, 13, 17, 3, 14, 21, 5, 5, 4, 6, 3, 13, 11, 18, 1, 17, 20, 19, 2, 20, 19, 26, 21, 4, 20, 21, 3, 24, 22, 14, 18, 17, 22, 4, 15, 17, 6, 14, 18, 6, 11, 21, 4, 24, 18, 28, 18, 16, 0, 21, 30, 28, 11, 67\}.

In the context of Bayesian estimation, $c_\Delta$ is the credibility of the region, if $f(\rho)$ is the posterior distribution.

When the constraint is strictly imposed throughout, one could make use of a Choiley decomposition to check for a semi-positive-definite and hermitian matrix. Then, the positivity of the smallest eigenvalue of the matrix corresponding to $\rho$ is used to impose the soft constraint via the indicator function.

The covariance matrix is more accurately estimated when the sample is larger.

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Whereas the “accelerated hit-and-run” algorithm used in [31][32] assumes that the $A$-regions have ellipsoidal shape, which is only justified for $A \lesssim 1$, no such assumptions enter the SCMC algorithm.

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Generally, the number of MC iterations, $N_{MC}$, is chosen according to [41].

$$N_R = \left\lfloor \log_{1-\alpha}(r) \right\rfloor,$$

where $N_R$ is the required number of MC steps such that there is a $(1 - q)$ probability that each point is moved at least once during the MC procedure which has an acceptance rate of $\alpha$. To put in some numbers for illustration, $N_{MC} = 17$ when $q = 0.01$ and $\alpha = 0.25$ for a typical MHMC, which has a most efficient acceptance rate of 0.234 [62], and we choose $N_{MC} = 15$ for convenience throughout the paper.

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