OVERCOMING THE NUMERICAL SIGN PROBLEM IN WIGNER DYNAMICS VIA PARTICLE ANNIHILATION

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Abstract. The infamous numerical sign problem poses a fundamental obstacle to long-time stochastic Wigner simulations in high dimensional phase space. Although the existing particle annihilation via uniform mesh (PAUM) significantly alleviates the sign problem when dimensionality $D \leq 4$, the setting of regular grids gives rise to another challenge in data storage when $D \geq 6$ due to the curse of dimensionality. In this paper, we propose to use a recently developed adaptive particle annihilation, termed sequential-clustering particle annihilation via discrepancy estimation (SPADE), to overcome the numerical sign problem. SPADE consists of adaptive clustering of particles via controlling their number-theoretic discrepancies and independent random matching in each group, and may learn the minimal amount of particles that can accurately capture the oscillating nature of the Wigner function. Combining SPADE with a recently proposed variance reduction technique via the stationary phase approximation, we make the first attempt to simulate the transitions of hydrogen energy levels in 6-D phase space, where the feasibility of PAUM with sample sizes about $10^9$ to $10^{10}$ has also been explored as a comparison.

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1. Introduction. Numerical methods of Wigner quantum dynamics\(^1\) have burgeoned with a vast number of developments during the past decades and allow a wide spectrum of applications in semiconductor devices and nano-materials\(^2–8\) as well as in studying typical quantum phenomena, such as the quantum decoherence,\(^9\) double-slit interference\(^10\) and many-body quantum effects.\(^11–13\) Nowadays, deterministic Wigner solvers are able to produce highly accurate results for 4-D problems due to their solid mathematical theory and concise guiding principle.\(^10, 13\) Meanwhile, stochastic particle methods, including the Wigner Monte Carlo,\(^4, 6, 7, 14\) the random cloud model\(^15, 16\) and the Wigner branching random walk (WBRW),\(^17–19\) can also resolve the quantum dynamics in 4-D phase space and have the potential to overcome the curse of dimensionality due to its theoretical convergence order $N^{-1/2}$ with $N$ the initial effective particle number (sample size), regardless of dimensionality $D$.

Despite these huge achievements, until very recently have few results been reported for stochastic Wigner simulations of realistic physical models in 6-D or higher dimensional phase space, even though they are of great importance in applications such as investigating the transition of energy levels and the relaxation of excited states to lower energy states,\(^20\) as well as the visualization of quantum states.\(^21–23\) The formidable obstacle to the particle-based Wigner simulation is the exponential growth of both particle number and stochastic variance, known as the notorious numerical sign problem.\(^19, 24, 25\) In our preceding work,\(^19\) we have pointed out that the sign problem is induced by the Hahn-Jordan decomposition of the pseudodifferential operator ($\Psi$DO), so that it is inherited in the oscillatory nature of nonlocal quantum interactions as well as the possible negative values of the Wigner function.

Although it is believed to be NP-hard in general,\(^26, 27\) the numerical sign problem can be significantly alleviated by fully utilizing the near-cancellation of positive and
negative particle weights. One approach directly aims at the reduction in the variances of the stochastic solutions, including the weighted-particle setting in the WBRW,\textsuperscript{17} the stationary phase approximation (SPA)\textsuperscript{19} and the semiclassical approximation.\textsuperscript{28} Another approach is the particle resampling, including the particle annihilation via uniform mesh (PAUM)\textsuperscript{4,6,7,14,17} and the particle resampling by filtering out the high-frequency components.\textsuperscript{29} Combining these together allows the long-time simulations of the Wigner dynamics in device applications, such as the coherent transport in resonant tunneling diode.\textsuperscript{4,6} Its numerical accuracy for $D \leq 4$ has already been benchmarked by careful comparisons with the reference solutions obtained by highly accurate deterministic solvers.\textsuperscript{17,19,30} However, it is still difficult to extend PAUM to $D \geq 6$ because the efficiency of uniform mesh is undermined by the curse of dimensionality. On one hand, the number of bins in a uniform mesh grows exponentially in $D$. On the other hand, the required sample size for given accuracy also grows with the increasing number of bins.\textsuperscript{18,31}

The goal of this paper is to utilize an adaptive strategy, dubbed the sequential-clustering particle annihilation via discrepancy estimation (SPADE),\textsuperscript{32} to ameliorate the curse of dimensionality, partially borrowing the pioneering idea in the non-parametric density estimation based on discrepancy estimation.\textsuperscript{33} SPADE consists of adaptive clustering of particles via controlling their number-theoretic discrepancies and independent random matching among positive and negative particles in each group. A thorough numerical study will demonstrate that SPADE achieves an efficient annihilation for a wide range of sample sizes and is capable to recover the “bottom line structure” pointed out in,\textsuperscript{30} which describes the minimal amount of particles that can accurately capture the oscillating nature of the Wigner function. Therefore, SPADE overcomes the essential drawbacks in the setting of regular mesh and may pave the way for realistic simulations, especially many-body problems in high-dimensional phase space. It deserves to mention that the calculation of the number-theoretic discrepancy of a sequence, as a pivotal step in adaptive clustering, is NP-hard in nature.\textsuperscript{34} In a sense, SPADE resolves the numerical sign problem by seeking efficient heuristic approximations to another NP-hard problem.

With the developments of particle annihilation and WBRW-SPA,\textsuperscript{19} we make the first attempt to simulate the electron dynamics of a hydrogen atom in 6-D phase space. Since the analytical formula of the Wigner function is not even known for the $1s$ state of the Hydrogen atom, we start from the Gaussian-type orbitals, which are ubiquitously adopted in quantum chemistry,\textsuperscript{35,36} and obtain the corresponding Gaussian approximation to the Hydrogen Wigner functions. Simulating the dynamics of a superposition of $1s$ and $2s$ states not only provides a visualization of the transitions between two bounded states, but also produces the differences of energy levels through power spectral analysis. As a comparison, we try to confront the challenges in both data storage and simulation cost of PAUM and demonstrate its feasibility in 6-D phase space with the help of parallel and distributed computing. In our experiment, a uniform mesh of size $60^3 \times 80^3 \approx 1.1059 \times 10^{11}$ is adopted for particle annihilation and required sample size ranges from $10^9$ to $10^{10}$ to ensure its efficiency.

The rest of paper is organized as follows. Section 2 briefly reviews the particle method and the numerical sign problem. Section 3 focuses on the design of SPADE. Section 4 derives the Gaussian Wigner function and the corresponding sampling strategy, as well as the WBRW-SPA model for the Coulomb system. Numerical results are reported in Section 5. Finally, conclusion and discussion are drawn in Section 6.
2. Particle generation and numerical sign problem. The Wigner function is defined by the Weyl-Wigner transform of the wavefunction $\phi(x, t)$, with $\phi^*$ the complex conjugate of $\phi$,

$$f(x, k, t) = \frac{1}{(2\pi)^3} \int_{\mathbb{R}^3} \phi^*(x - \frac{y}{2}, t)\phi(x + \frac{y}{2}, t)e^{-ik\cdot y}dy.$$  \hspace{1cm} (2.1)

In principle, the Wigner dynamics allows to investigate the excited states without any a priori knowledge of eigenstates.\(^{20}\) It requires to solve the Wigner equation, a time-dependent partial integro-differential equation,

$$\frac{\partial}{\partial t} f(x, k, t) + \frac{\hbar k}{m} \cdot \nabla_x f(x, k, t) = \Theta_V[f](x, k, t),$$  \hspace{1cm} (2.2)

where $k/m$ is short for $(k_1/m_1, \ldots, k_N/m_N)$, $m_i$ is the mass of the $i$-th body, $\hbar$ is the reduced Planck constant and $\Psi DO$ reads as

$$\Theta_V[f](x, k, t) = \frac{1}{\hbar(2\pi)^n} \int_{\mathbb{R}^{2n}} e^{-i(k-k')\cdot y}(V(x + \frac{y}{2}) - V(x - \frac{y}{2}))f(x, k', t)dydk'.$$

Formally, Eq. (2.2) has a plane wave expansion of the quantum observables,

$$\langle A \rangle(t) = \langle A_W, f(t) \rangle = \sum_{n,m=0}^{\infty} a_{n,m}e^{-\frac{\pi E_n - E_m t}{\gamma}} \langle A_W, f_{n,m} \rangle,$$

where $A_W$ is the Weyl symbol of the quantum operator $A$,

$$\langle \varphi, f \rangle = \iint_{\mathbb{R}^{2n}} \varphi(x, k) f(x, k) dx dk,$$

$E_n$ are $n$-th energy level of the hydrogen atom and $f_{n,n}$ and $f_{n,m}(m \neq n)$ are the Wigner functions corresponding to $n$-th eigenstate and $(n, m)$-correlation, respectively. In this manner, the differences in energy level pairs $(E_n, E_m)$ can be obtained via a direct spectrum analysis of $\langle A \rangle(t)$.

The particle simulation of the deterministic Wigner equation (2.2) is based on its stochastic representation, which interprets the formal Neumann series expansion as the expectation of stochastic trajectories over Poisson jumps.\(^{7,14-17,19}\) From the mathematical perspective, it utilizes the fact that for any test function $\varphi \in L^2(\mathbb{R}^{2n})$ and initial condition $f_0 \in L^2(\mathbb{R}^{2n})$, there exists a branching random walk model $X_t$ and two constants $C_1 > 0, C_2 > 0$ such that\(^{19}\)

$$\mathbb{E}(X_t, f_0) = \langle \varphi, f(t) \rangle, \quad \mathbb{E}(\langle X_t, f_0 \rangle - \langle \varphi, f(t) \rangle)^2 \leq C_1 \exp(C_2 t).$$  \hspace{1cm} (2.5)

Although the expectation of the stochastic model solves the Wigner equation, both the stochastic variance and particle number (taking $\varphi = 1$) grow exponentially, thereby posing a formidable limitation to the particle method especially for long-time simulations. Such phenomenon is called the numerical sign problem, which stems from the near-cancellation of positive and negative weights in numerical integrations of oscillatory functions.\(^{24,26}\) Because of a large variance of estimator, sample size must be large enough to obtain reliable results within a small relative uncertainty.

To alleviate the sign problem, we have recently proposed a variance reduced model, termed the WBRW-SPA.\(^{19}\) It utilizes SPA to capture the decay of high-frequency component of $\Psi DO$, giving rise to another WBRW model and a constant $\alpha^* < 1$ such that

$$\mathbb{E}(X_t, f_0) = \langle \varphi, f(t) \rangle + \mathcal{O}(\lambda_0^{-1}), \quad \mathbb{E}(\langle X_t, f_0 \rangle - \langle \varphi, f(t) \rangle)^2 \lesssim \exp(\alpha^* C_2 t).$$  \hspace{1cm} (2.6)
WBRW-SPA suppresses the exponential growth of both particle number and stochastic variances compared with all existing stochastic algorithms, at the cost of introducing a small asymptotic error term $O(\lambda^{-1})$, thereby significantly improving the numerical accuracy.\(^{19}\)

By taking average of independent realizations of WBRW-SPA, we obtain the following estimator

\[
\langle \varphi, f(t) \rangle \sim \langle \varphi, \mu \rangle, \quad \mu = \frac{1}{N_\alpha} \sum_{i=1}^{P} \delta(x_i^+, k_i^+) - \frac{1}{N_\alpha} \sum_{i=1}^{M} \delta(x_i^-, k_i^-),
\]

where $S^+ = \{(x_i^+, k_i^+)\}_{i=1}^{P}$ and $S^- = \{(x_i^-, k_i^-)\}_{i=1}^{M}$ are positive and negative particles, carrying opposite particle weight $\pm 1$, respectively. The effective particle number, or the normalizing constant, is $N_\alpha = P - M$. In other words, the particle method seeks an empirical sign measure $\mu$ to approximate the Wigner function in the weak sense.

3. Particle annihilation. The numerical sign problem cannot be completely surmounted by SPA because it is rooted in the nonlocal nature of the low-frequency component of $\Psi_{DO}$. Moreover, it is more probably to be aggravated as the dimensionality (system size) increases due to the enrichment of fine structures, such as the alternation of local maxima and minima, in the Wigner function.

To further alleviate the sign problem, especially for long-time simulations, particle annihilation turns out to be an indispensable operation. For a given empirical signed measure of the form in Eq. (2.7), particle annihilation intends to remove $N_A$ positive particles from $S^+$ and $N_A$ negative ones from $S^-$ and obtain another empirical signed measure $\nu$,

\[
\nu = \frac{1}{N_\alpha} \sum_{i=1}^{P-N_A} \delta(\tilde{x}_i^+, \tilde{k}_i^+) - \frac{1}{N_\alpha} \sum_{i=1}^{M-N_A} \delta(\tilde{x}_i^-, \tilde{k}_i^-).
\]

Here $(\tilde{x}_i^+, \tilde{k}_i^+)$ can be either chosen as a subset of $S^+$, or be generated by certain operations of particles in $S^\pm$. The target of particle annihilation is to control the error function $E(\varphi) = |\langle \varphi, \mu \rangle - \langle \varphi, \nu \rangle|$ for suitable test functions $\varphi$. It is expected to annihilate two kinds of particles carrying opposite importance weights and cancelling out their contributions within a reasonable numerical accuracy. For this reason, the particle annihilation is also known as particle cancellation or particle resampling in some occasions.\(^{29}\)

The prototype PAUM\(^4,6,14\) borrows the essential idea from the histogram statistics.\(^{31}\) In spite of its simplicity and easy implementation, the efficiency of the uniform mesh deteriorates sharply as the dimension increases, known as the curse of dimensionality. An adaptive particle annihilation algorithm is introduced to overcome the severe limitation of regular mesh.\(^{32}\) We will focus on the intuition, design and implementation of SPADE, and a thorough numerical comparison between PAUM and SPADE is left in Section 5.

3.1. PAUM: particle annihilation via uniform mesh. Suppose the computational domain is a rectangular bin $\mathcal{X} \times \mathcal{K} = \prod_{i=1}^{n}[x_{\text{min}}^{(i)}, x_{\text{max}}^{(i)}] \times \prod_{j=1}^{n}[k_{\text{min}}^{(j)}, k_{\text{max}}^{(j)}]$. A simple idea is to utilize a uniform mesh for partitioning the computational domain $\mathcal{X} \times \mathcal{K} = \bigcup_{k=1}^{K} \mathcal{Q}_k$, where $\mathcal{Q}_k = \mathcal{X}_{i_1} \times \cdots \times \mathcal{X}_{i_n} \times \mathcal{K}_{j_1} \times \cdots \times \mathcal{K}_{j_n}$ is the tensor product
of rectangular bins

\( X_i = [x^{(m)}_{\min} + (i_m - 1)\Delta x_m, x^{(m)}_{\min} + i_m\Delta x_m], \quad m = 1, \ldots, n, \) \hfill (3.2)

\( K_j = [k^{(m)}_{\min} + (j_m - 1)\Delta k_m, k^{(m)}_{\min} + j_m\Delta k_m], \quad m = 1, \ldots, n, \) \hfill (3.3)

and \( \Delta x_m \) and \( \Delta k_m \) are spatial and momental spacings in the \( m \)-dimension, respectively. PAUM suggests to use a piecewise constant function \( p(x, k) \) to approximate the Wigner function \( f(x, k, t) \),

\[
p(x, k) = \sum_{k=1}^{K} \frac{P_k - N_k}{N_\alpha} \cdot \frac{1}{\text{vol}(Q_k)},
\]

where \( P_k \) and \( M_k \) are counts of the positive and negative particles in the bin \( Q_k \), respectively, and \( \text{vol}(Q_k) \) is the Lesbegue measure of \( Q_k \). Thus the particles carrying opposite signs are eliminated directly.\(^7\)

Details of resampling from \( p(x, k) \) and the theoretical analysis can be found in\(^18\) and omitted here for brevity. Here we only point out the pros and cons of PAUM.

1. PAUM requires to store a uniform mesh instead of particles. Once particles are counted, they can be destroyed. This makes PAUM convenient for parallel and distributed computing.
2. The uniform mesh essentially provides an approach to clustering particles into \( K \) groups and the error can be diminished by decreasing the size of bins.
3. PAUM is bothered by the curse of dimensionality since \( K \) grows exponentially as dimensionality \( D \) increases. For instance, in 4-D space, it requires 384MB to store an integer-valued matrix of partition level \( K = 100^4 \). However, in 6-D space, it requires 415GB to store an integer-valued matrix of partition level \( K = 60^{3} \times 80^{3} \), so that PAUM might not be extended to \( D > 6 \).
4. The efficiency of PAUM is sensitive to the partition level \( K \) and the sample size \( N_\alpha \) (normalizing constant in the estimator Eq. (2.7)). In general, it works well only when \( K \) is comparable to \( N_\alpha \); otherwise it might not be able to annihilate most of particles or to control the stochastic variances due to the severe “overfitting” phenomenon.\(^{18,31}\)

This paper makes the first trial to explore the feasibility of PAUM in 6-D space. In our experiment, when \( K = 60^{3} \times 80^{3} \), \( N_\alpha \) must be chosen to be about \( 10^9-10^{10} \) to maintain its efficiency.

### 3.2. SPADE: an adaptive particle annihilation.

The relevant idea to overcome the curse of dimensionality in SPADE is to replace the uniform mesh by an adaptive one via a divide-and-conquer strategy.\(^32\) First, it seeks an adaptive partition of \( X \times K \) via the sequential binary splitting and controls the number-theoretic discrepancies of points in each group. Once an adaptive partition \( X \times K = \bigcup_{k=1}^{K} Q_k \) is obtained, it also divides the positive and negative particles into \( K \) groups, namely, \( S^+ = \bigcup_{k=1}^{K} S^+_k \), \( S^- = \bigcup_{k=1}^{K} S^-_k \). Second, it seeks a matching between the positive and negative particles in the same group independently and the annihilation can be realized by removing the matched pairs. The convergence issue of SPADE has been validated in our recent work,\(^32\) utilizing the Koksma-Hlawka inequality and concentration inequalities for sampling with or without replacement.

Now we illustrate the detailed implementation of SPADE via a recursive binary splitting. A binary partition \( P \) on a domain \( Q \) is the collection of sub-rectangles whose union is \( Q \). Starting with \( P_1 = \{Q\} \) at level 1, for \( P_K = \{Q_1, \ldots, Q_K\} \) at level
Then the particles are divided into 6 groups. The other point is the choice of dimension and node to split. A rule of thumb for particle annihilation problem is to dig out the possible nodal points in the hypersurfaces. To this end, we can adopt the following strategy. For the bin $Q_k$ is $P_0 = (Q_1, \ldots, Q_6)$, and each bin ceases to be split when both positive and negative particles satisfy the discrepancy bounds (3.9) simultaneously; otherwise it will be split further into two parts. In the mean time, the particles are divided into 6 groups.

The other point is the choice of dimension and node to split. A rule of thumb for particle annihilation problem is to dig out the possible nodal points in the hypersurfaces. To this end, we can adopt the following strategy. For the bin $Q_k$ is $[a_{k,1}, b_{k,1}] \times \cdots \times [a_{k,2n}, b_{k,2n}]$, we can pick up $j$-th dimension and $m$ equidistant points in the interval $[a_{k,j}, b_{k,j}]$, say, $\zeta = a_{k,j} + \frac{1}{m}(b_{k,j} - a_{k,j}), l = 1, \ldots, m - 1$, then select the node $\zeta$ from $(m - 1) \times 2n$ choices (typically, $m = 2, 4, 8$). Such node splits the bin $Q_k$ into $Q_k^{(1)}$ and $Q_k^{(2)}$,

$$Q_k^{(1)} = \prod_{i=1}^{j-1} [a_{k,i}, b_{k,i}] \times [a_{k,j}, \zeta] \times \prod_{i=j+1}^{2n} [a_{k,i}, b_{k,i}],$$

$$Q_k^{(2)} = \prod_{i=1}^{j-1} [a_{k,i}, b_{k,i}] \times [\zeta, b_{k,j}] \times \prod_{i=j+1}^{2n} [a_{k,i}, b_{k,i}],$$

with $P_k^{(1)}$ and $M_k^{(1)}$ are counts of positive and negative particles in $Q_k^{(1)}$, respectively. Then the choice of $\zeta$ is to attain the maximal value of the following maximal gap:

$$\max \left( \frac{P_k^{(1)}}{P_k} - \frac{\text{vol}(Q_k^{(1)})}{\text{vol}(Q_k)}, \frac{M_k^{(1)}}{M_k} - \frac{\text{vol}(Q_k^{(1)})}{\text{vol}(Q_k)} \right).$$
In practice, we have found that the maximal gap works better for \( D \leq 6 \), while the difference gap introduced in\(^{12}\) performs better for \( D > 6 \).

The flowchart of SPADE is given in Algorithm 1. SPADE tries to convert the numerical sign problem into calculation of the star discrepancy, which is still a NP-hard problem and essentially difficult to solve exactly. Fortunately, the star discrepancy can be approximated by some heuristic algorithms for the integer optimization. Here we adopt the improved version of threshold accepting algorithm (TA-improved algorithm) with probabilistic sampling proposed in,\(^{14}\) which gives an efficient approximation to the star discrepancy in moderately large dimension (\( D \leq 60 \)).

Now we make a comparison between SPADE and PAUM.

1. In contrast to PAUM, SPADE requires to store all the particles in a \((P+M) \times 2n\) real-valued matrix and an adaptive partition in a \(K \times (4n+2)\) real-valued matrix (including upper and lower bounds of bins and counts of particles). Since \( K \) is usually smaller than \( P+M \), the data storage in SPADE scales linearly with \( D = 2n \), thereby overcoming the curse of dimensionality.

2. The binary partition in SPADE is refined automatically according to the concentration and sparsity of point distributions. A 2-D visualization of a uniform mesh and an adaptive partition is presented in Fig. 2. One can readily see both PAUM and SPADE are able to recover the Wigner function from discrete samples, while the adaptive partition tends to be refined in the region where the samples are concentrated and ceases to be split further when points are sparsely distributed. This manifests the adaptivity of SPADE.

3. SPADE can monitor an appropriate partition level \( K \) for given effective sample size \( N_\alpha \) and the parameter \( \theta \). Since \( K \) is usually smaller than \( N_\alpha \), SPADE
Algorithm 1 SPADE: An adaptive particle annihilation

**Input parameters:** The domain \( Q \), the positive particles \( S^+ \), the negative particles \( S^- \), the normalizing constant \( N_\alpha \) and the parameter \( \vartheta \).

**Clustering:** Start from \( K = 1 \), \( Q_1 = Q \), \( P = \{Q_1\} \), \( P' = \emptyset \), \( S^+_1 = S^+ \), \( S^-_1 = S^- \).

while \( P \neq P' \) do
  \( P' = P \)
  for all \( Q_k \) in \( P' \) do
    \( P \leftarrow P \setminus Q_k \)
    \( S^+ \leftarrow S^+ \setminus S^+_k \), \( S^- \leftarrow S^- \setminus S^-_k \)
    Calculate the star discrepancies \( \Delta_{P_k}^*(S^+_k) \) and \( \Delta_{M_k}^*(S^-_k) \)
    if \( \Delta_{P_k}^*(S^+_k) > \vartheta \sqrt{N_\alpha} \) or \( \Delta_{M_k}^*(S^-_k) > \vartheta \sqrt{N_\alpha} \) then
      \( K \leftarrow K + 1 \)
      Choose a node \( \zeta \) to attain the maximum of Eq. (3.11)
      Divide \( Q_k \) into \( Q_k^{(1)} \) and \( Q_K \leftarrow Q_k^{(2)} \) as given in Eq. (3.10)
      Divide the pointsets: \( S^+_k \rightarrow S^+_k \cup S^+_K \)
      Update the partition and particles: \( P \leftarrow P \cup Q_k \cup Q_K \), \( S^+ \leftarrow S^+ \cup S^+_k \cup S^+_K \)
    else
      \( P \leftarrow P \cup Q_k \), \( S^+ \leftarrow S^+ \cup S^+_k \)
    end if
  end for
end while
return \( Q = \bigcup_{k=1}^K Q_k \), \( S^+ = \bigcup_{k=1}^K S^+_k \), \( S^- = \bigcup_{k=1}^K S^-_k \)

**Matching:** For \( k \)-th group, when \( P_k \geq M_k \), seeking a random matching from \( S^-_k \) to \( S^+_k \). Otherwise, seeking a random matching from \( S^+_k \) to \( S^-_k \). The random matchings are mutually independent.

**Annihilation:** Remove the paired particles in each group.

can work even when \( N_\alpha \) is relatively small. In a sense, it alleviates the over-fitting problem.

4. The error bound of SPADE converges to 0 as \( N_\alpha \rightarrow \infty \) and it is also influenced by the parameter \( \vartheta \), the partition level \( K \) and the total variation of test function in the sense of Hardy and Krause.\(^{32}\)

4. Particle simulation of the Coulomb system. Excited states are those quantum states of an atom or a molecule with more energy than the ground state. Understanding of excited states plays an important role in spectroscopy and also lies at the heart of photochemistry. However, most of quantum Monte Carlo methods that utilize the formal similarity of the Schrödinger equation to a diffusion equation aim at producing a reliable estimate of ground state energy and only a few have been extended to excited states.\(^{37}\) By contrast, the stochastic Wigner simulation may not only provide a natural way to investigate energy transitions and but also allow a visualization of excitation of electron in phase space.

The hydrogen atom is a fundamental quantum molecular system composed of one proton and one electron interacting under the attractive Coulomb potential,

\[
V(x) = -\frac{1}{|x - x_A|}, \quad x \in \mathbb{R}^3.
\]

Suppose the proton is fixed at \( x_A \) under the Born-Oppenheimer approximation, one
can obtain the eigenvalues of quantum Hamiltonian operator in atomic units ($\hbar = m = e = 1$) reads as

\[(4.2) \quad (-\Delta_x + V(x)) \phi_n(x) = E_n \phi_n(x), \quad E_n = -\frac{1}{2n^2}, \quad n = 1, 2, \ldots.\]

However, the exact Wigner function even for 1s state cannot be written in an analytical form.\textsuperscript{38} Fortunately, the sets of Gaussian-type functions have been frequently employed as the basis function in molecular electronic structure calculation owing to the simplicity of evaluating the required integrals,\textsuperscript{35} thereby serving as the nexus of both the Wigner and the wave function approaches. Despite that it is usually difficult to obtain the Wigner functions of exact eigenstates, the Gaussian Wigner function (GWF) provides a good approximation with proper symmetry and produces a physically correct picture.

4.1. Gaussian Wigner function. In general, the atomic orbitals are presented by Gaussian-type orbitals (GTOs) $\phi_j(x)$ with origin at point $x_j$ in space and variable exponent $a_j$ and have the following general form:\textsuperscript{35,36}

\[(4.3) \quad \phi(x) = \frac{1}{N} \sum_{j=1}^{N_{\text{basis}}} c_j \phi_j(x) = \frac{1}{N} \sum_{j=1}^{N_{\text{basis}}} c_j \left(\frac{2a_j}{\pi}\right)^{3/4} \exp(-a_j|x|^2), \quad x \in \mathbb{R}^3.\]
Substituting Eq. (4.3) into (2.1) directly yields GWF

(4.4)
\[ f(x, k) = \frac{1}{N^2} \sum_{j=1}^{N^2} c^2_j f_{jj}(x, k) + \frac{1}{N^2} \sum_{j<k} c_j c_k (f_{jk}(x, k) + f_{kj}(x, k)) \]
\[ = \sum_{j=1}^{N^2} \frac{c^2_j}{N^2} g_{jj}(x, k) + \sum_{j<k} \frac{2c_j c_k}{N^2} g_{jk}(x, k) \cos \left( \frac{2(a_j (x - x_j) - a_k (x - x_k)) \cdot k}{a_j + a_k} \right), \]

where

(4.5) \[ g_{jk}(x, k) = \left[ \frac{(a_j + a_k) \pi}{4a_j a_k} \right]^{3/2} \left[ \frac{4a_j a_k}{(a_j + a_k)^2 \pi^2} \right]^{3/2} \phi \left( \frac{4a_j a_k}{a_j + a_k} \right) \left( x - \frac{x_j + x_k}{2} \right)^2 - \frac{|k|^2}{a_j + a_k}. \]

One can readily realize that the possible negative values of GWF come from the phase factors in off-diagonal elements, which exactly reflects the correlation between two different Gaussian-type orbitals. The above derivation can be straightforwardly generalized to the superposition states and the Slater-determinant correspondences can be built by the tensor product of single-body GWFs.

Sampling according to Eq. (4.4) can be realized by the following population sampling algorithm. It is suggested to choose the instrumental probability density \( f_I \) by omitting the phase factors, say,

(4.6) \[ f_I(x, k) = \frac{1}{S} \sum_{s=1}^{S} f_s(x, k) \]
with \( S = N_{basis}(N_{basis} + 1)/2 \), \( f_s(x, k) \) are normalized Gaussian functions \( g_{jk}(x, k) \), \( 1 \leq i \leq j \leq N_{basis} \). Now we illustrate the population sampling in Algorithm 2, where \( \lfloor p \rfloor \) denotes the integer part of \( p \), and coefficients of different states are put in Appendix (see Table 4).

**Algorithm 2** Population sampling from the GWF \( f(x, k) \)

**Input parameters**: Initial effective particle number (sample size) \( N_\alpha \).
1. Choose a random integer \( s \) uniformly from the set \( \{1, \ldots, S\} \).
2. Draw a sample \( (x_\alpha, k_\alpha) \) from \( f_s(x, k) \).
3. Compute the ratio \( p = |f(x_\alpha, k_\alpha)|/f_I(x_\alpha, k_\alpha) \) and weight \( w = \frac{f(x_\alpha, k_\alpha)}{f_I(x_\alpha, k_\alpha)} \).
4. Draw a uniform random number \( u \) in \( [0, 1] \).
5. If \( p - \lfloor p \rfloor < u \), we preserve \( p - \lfloor p \rfloor \) copies of \( (x_\alpha, k_\alpha) \) endowed with signed weight \( w \). Otherwise, we preserve \( p - \lfloor p \rfloor \) copies of \( (x_\alpha, k_\alpha) \).
6. Go back to Step 1 and repeat the procedure \( N_\alpha - 1 \) times.

### 4.2. WBRW-SPA for 3-D Coulomb interaction

In particular, for the attractive Coulomb potential (4.1), the corresponding \( \Psi \)DO turns out to be

(4.7) \[ \Theta_V[f](x, k, t) = \int_{\mathbb{R}^3} e^{i z(x) \cdot k'} \psi(k') (f(x, k - \frac{k'}{2}, t) - f(x, k + \frac{k'}{2}, t)) d\mathbf{k}' \]
with \( z(x) = x - x_A \) and the amplitude function

(4.8) \[ \psi(k) = \frac{1}{i \hbar (2\pi)^3} \left( \frac{2\pi}{c_{3,1}} \right)^{1/2} |k|^2 \frac{1}{\hbar c_{3,1}} \frac{1}{|k|^2}, \quad c_{n,\alpha} = \pi^{n/2} \Gamma(\frac{n}{2}) \Gamma(\frac{n+\alpha}{2}). \]
The phase function depends on the spatial displacement \( \mathbf{x} - \mathbf{x}_A \), so that the integrand becomes more oscillated as the distance \(|\mathbf{x} - \mathbf{x}_A|\) increases. This coincides with our intuition as the quantum interaction should decay when two bodies are far apart.

In practice, \( \Psi_D \) of the form (4.7) poses the combined computational challenges of high oscillations and high dimension. To deal with these problems, we use SPA to capture the decay of its high-frequency component, yielding another operator \( \Theta_{(4.9)}^\lambda[f](\mathbf{x}, \mathbf{k}, t) \),

\[
\Theta_{(4.9)}^\lambda[f](\mathbf{x}, \mathbf{k}, t) = \Lambda^{\leq \lambda_0}[f](\mathbf{x}, \mathbf{k}, t) + \Lambda^{> \lambda_0}[f](\mathbf{x}, \mathbf{k}, t) + \Lambda^{> \lambda_0}[f](\mathbf{x}, \mathbf{k}, t),
\]

where \( \lambda_0 \) is specified filter and the low-frequency component under a spherical coordinate \( \mathbf{k}' = (r \cos \theta, r \sin \theta \cos \phi, r \sin \theta \sin \phi) \) reads that

\[
\Lambda^{\leq \lambda_0}[f](\mathbf{x}, \mathbf{k}, t) = \frac{1}{\hbar c_{3.1}} \int_{|\mathbf{k}'| \leq \lambda_0} \sin(|\mathbf{x} - \mathbf{x}_A|) \left( f(\mathbf{x}, \mathbf{k} - \frac{k'}{2}, t) - f(\mathbf{x}, \mathbf{k} + \frac{k'}{2}, t) \right) \mathrm{d}k',
\]

and the upper limit \( \Lambda^{> \lambda_0}[f](\mathbf{x}, \mathbf{k}, t) = 1 \) for \( |\mathbf{k}'| > \lambda_0 \).

Two principal terms in the asymptotic expansion of high-frequency component are

\[
\Lambda^{> \lambda_0}[f](\mathbf{x}, \mathbf{k}, t) = \pm \frac{4\pi}{\hbar c_{3.1}} \int_{r_0}^{r_{\max}} \sin(|\mathbf{z}(\mathbf{x})|) f(\mathbf{x}, \mathbf{k} + \frac{r \sigma_\star(\mathbf{x})}{2}, t) \mathrm{d}r, \quad \{k_0, \mathbf{z}(\mathbf{x}) \in \mathcal{K} \},
\]

where the critical point is parametrized by \( \sigma_\star(\mathbf{x}) = (\cos \theta^*, \sin \theta^* \cos \phi^*, \sin \theta^* \sin \phi^*) \),

\[
\theta^* = \text{atan}2(\sqrt{(x_2 - x_{A,2})^2 + (x_3 - x_{A,3})^2}, x_1 - x_{A,1}),
\]

\[
\phi^* = \text{atan}2(x_3 - x_{A,3}, x_2 - x_{A,2}).
\]

The wavenumber \( \mathbf{k} \) is truncated at \( \mathcal{K} \) and the upper limit \( r_{\max} \) satisfy \( r_{\max} \geq 2|\mathcal{K}| \).

A transparent physical interpretation of SPA is that the major contribution of the high-frequency quantum interaction comes from the nonlocal scattering along the line \( \mathbf{x} - \mathbf{x}_A \), and the weight decays as \( O(|\mathbf{z}(\mathbf{x})|^{-1}) \), while the contributions deviated from \( \mathbf{x} - \mathbf{x}_A \) almost cancel out.

The detailed implementation of WBRW-SPA for the 3-D attractive Coulomb potential is illustrated in Algorithm 3.

5. Particle simulation of 6-D Wigner quantum dynamics. With the above preparations, we begin to investigate the 6-D phase-space quantum dynamics of an electron in a hydrogen atom.

The first test explores the feasibility of PAUM for long-time Wigner simulation (up to \( t = 100 \text{a.u.} \)). With parallel and distributed computing (128 cores), it produces a reliable reference solution under a \( 60^3 \times 80^3 \) grid mesh and \( N_o = 10^9 \) with the computational time more than 20 days. Second, we will show that under the same computational platform and a relatively small sample size \( N_o = 1 \times 10^7 \), WBRW-SPA combined with SPADE, albeit with some loss of accuracy, is able to capture the transitions in energy levels, while the computational time can be shortened to about 13 hours (\( \vartheta = 0.005 \)) or 8 hours (\( \vartheta = 0.01 \)) and the requirement of data storage is dramatically reduced.

In order to further investigate how the parameter \( \vartheta \) in SPADE and sample size \( N_o \) influence the numerical accuracy, energy conservation, particle number and the
**Algorithm 3** Signed-particle WBRW-SPA for the Coulomb system

**Input parameters:** The initial time $t_0$ and final time $t_{f+1}$, the constant rate $\gamma_0$, the filter $\lambda_0$, $k$-domain $K$ and the upper band $r_{max}$.

**Sampling processes:** Suppose each particle in the branching particle system, carrying an initial weight $w$ either 1 or $-1$, starts at state $(x_0, k_0)$ at time $t_0$ and moves until $t_{f+1} = t_0 + \Delta t$ according to the following rules.

1. (Frozen) Generate a random $\tau \sim \gamma_0 e^{-\gamma_0 t}$. For a particle at $(x_0, k_0)$ at instant $t \in [t_0, t_{f+1}]$, if $t + \tau \geq t_{f+1}$, it becomes frozen at $(x + \hbar k_0(t_{f+1} - t)/m, k_0, (t_{f+1})$.

2. (Death) If $\tau < \Delta t$, the particle moves to $(x + \hbar k_0 \tau/m, k_0, t + \tau)$ and is killed.

3. (Branching) When the particle is killed, it produces at most three offsprings at states $(x_1, k_1, t + \tau)$, $(x_2, k_2, t + \tau)$ and $(x_3, k_3, t + \tau)$. The third offspring are produced at state $(x_3, k_3) = (x + \hbar k_0 \tau/m, k_0)$ with probability 1, carrying the weight $w$.
   
   (1) If $r < \lambda_0/|x_3 - x_A|$, generate random numbers $\varphi$ uniformly in $[0, \pi]$ and $\phi$ uniformly in $[0, 2\pi]$, yielding a vector $k'$. It produces two offsprings with probability $\Pr(1)$, $\Pr(2)$ at states $(x_1, k_1)$, $(x_2, k_2)$ endowed with updated weights $w_1$ and $w_2$, respectively.
   
   $\Pr(1) = \Pr(2) = \frac{2\pi^2 r_{max}}{\hbar c_{3,1} \gamma_0} |\sin((x_3 - x_A) \cdot k') \sin \phi|.
   
   $x_i = x_0 + \frac{\hbar k_0 \tau}{m}$, $k_i = k_0 + \frac{(-1)^i k'}{2}$, $i = 1, 2$.

   $w_i = w \cdot \frac{|\sin((x_i - x_A) \cdot k') \sin \phi|}{\sin((x_i - x_A) \cdot k') \sin \phi} \cdot \mathbb{I}(k_i \in K)$, $i = 1, 2$.

3. If $r \geq \lambda_0/|x_3 - x_A|$, it produces two offsprings with the probability $\Pr(1)$, $\Pr(2)$ at states $(x_1, k_1)$, $(x_2, k_2)$ endowed with updated weights $w_1$ and $w_2$, respectively.

   $\Pr(1) = \Pr(2) = \frac{4\pi r_{max}}{\hbar c_{3,1} \gamma_0} \frac{|\sin(r|x_3 - x_A|)|}{r|x_3 - x_A|}.
   
   $x_i = x_0 + \frac{\hbar k_0 \tau}{m}$, $k_i = k_0 + \frac{(-1)^i r \sigma(x_i)}{2}$, $i = 1, 2$.

   $w_i = w \cdot \frac{|\sin(r|x_i - x_A|)|}{\sin(r|x_i - x_A|)} \cdot \mathbb{I}(k_i \in K)$, $i = 1, 2$.

4. (Independence) The offsprings continue to move independently.

**Termination condition:** All particles in the branching particle system are frozen.

partition level $K$, we provide a series of benchmarks on SPADE, as well as some criteria to choose appropriate parameters in the simulations.

All the simulations performed via our own Fortran implementations run on the High-Performance Computing Platform of Peking University with the platform: 2*Intel Xeon E5-2697A-v4 (2.60GHz, 40MB Cache, 9.6GT/s QPI Speed, 16 Cores, 32 Threads) with 256GB Memory $\times$ 8.
5.1. Performance of PAUM. Here we use the atomic unit and \( x_A = 0 \).

**Parameters for particle generation:** A superposition of 1s state and 2s state is considered,

\[
\phi(x) = \frac{1}{\sqrt{2}} \phi_{1s}(x) + \frac{1}{\sqrt{2}} \phi_{2s}(x).
\]

The Wigner function is

\[
f(x, k) = \frac{1}{4} (f_{1s, 1s}(x, k) + f_{2s, 2s}(x, k) + 2f_{1s, 2s}(x, k)).
\]

We use 4 Gaussian basis to approximate 1s orbital and 7 Gaussian basis to approximate 2s orbital with coefficients given in Table 4 (see Appendix), so that the Wigner function is represented by a linear combination of \( 11 \times (11 + 1)/2 = 66 \) Gaussian Wigner functions. Algorithm 2 is adopted for sampling the initial Gaussian Wigner function. A visualization of the Wigner function for 1s-2s superposition state is shown in Fig. 3. A heavy tail in \( k \)-space is observed for the Wigner function, which is a remarkable feature of the 2s state. Actually, GWF is essentially a mixed quantum state and occupies higher energy levels with certain probability. In order to obtain an accurate solution, as well as to ensure the efficiency of resampling, \( N_\alpha \) is set to be \( 1 \times 10^9 \) so that the initial total particle number is \( P + M = 2.38 \times 10^9 \) and the initial effective particle number is \( N_\alpha = 1 \times 10^9 \).

The signed-particle WBRW-SPA is adopted for evolving the Wigner dynamics until the final time \( t_{fin} = 100 \) a.u., with the auxiliary function \( \gamma_0 = 50 \) and the filter of wave number in SPA \( \gamma_0 = 4 \). The growth rate of particle number is about \( e^{1.16t} \) for WBRW-SPA, which is much smaller than the rate \( e^{1.88t} \) when SPA is absent. However, the total particle number will be soon doubled when \( t = 0.6 \) a.u. and reach a hundredfold when \( t = 3.97 \) a.u. Without particle annihilation, the growth rate attains \( 2.39 \times 10^{50} \) at \( t_{fin} = 100 \) a.u., and so is the stochastic variance! This clearly demonstrates the curse of numerical sign problem.

**Parameters for PAUM:** The particle annihilation occurs every 1 a.u., that is, the time in divided into 100 intervals \( 0 = t_0 < t_1 < \cdots < t_{100} = t_{fin} \) with \( t_{t+1} - t_t = \Delta t = 1 \) a.u. The domain is \( \mathcal{X} \times \mathcal{K} = [-9, 9]^3 \times [-4, 4]^3 \) and we adopt a 60 \( \times 60 \times 60 \times 80 \times 80 \times 80 \) uniform mesh on \( \mathcal{X} \times \mathcal{K} \), so that the partition level \( K \approx 1.1059 \times 10^{14} \). In computation, we need to store two integer-valued matrice of such size and each one requires the memory about 414.72 GB (the total memory requirement is about 1.2 TB). No specified boundary condition is adopted and the particles moving outside the domain are not counted.

**Parallelization:** We use 128 threads in the simulations. The data are distributed into 8 nodes and each node provides 16 cores. We adopt a domain decomposition strategy by dividing \( \mathcal{X} \) evenly in each dimension of the spatial space, that is, it is decomposed into \( 2 \times 2 \times 2 = 8 \) parts. The data in different nodes are transferred via the Message Passing Interference (MPI) standard and the parallelization in each node is realized by the OpenMP library. This can significantly reduce the communication time and achieve a balance on the overload.

**Snapshots:** To demonstrate the dynamics of electrons, we take snapshots of the Wigner function on \((x_1, k_1)\) plane from \( t = 2 \) a.u. to \( t = 100 \) a.u., as shown in Fig. 3. At first stage, negative values of the Wigner function tend to be concentrated around \((x_1, k_1) = (4, 0.5)\) and \((x_1, k_1) = (-4, -0.5)\), indicating that particles in \( x_1 > 0 \) (\( x_1 < 0 \)) are forbidden to have certain positive (negative) momentum. The Wigner function rotates around the central area and presents an oscillating pattern, which
visualizes the transitions between energy levels. The distribution begins to spread in spatial coordinate due to the advection mechanism so that the electron has a certain probability to jump to higher energy levels, while certain velocities (wave numbers) are forbidden by the negative values of the Wigner function. Finally it arrives at a symmetric state ($t = 100\, \text{a.u.}$). A double-peak structure is presented after $t = 50\, \text{a.u.}$

**Energy conservation:** Energy conservation is an importance feature of a conservative quantum system and serves as an indicator for numerical accuracy. Thus we record the numerical energy in Fig. 4(a). One can see the fluctuation of energy at
each time step. A small jump of energy occurs when resampling is performed. After a long time simulation, the energy changes from $-0.314 \text{ a.u.}$ to $-0.157 \text{ a.u.}$ The 50% loss of energy seems to coincide with 42.6% of total mass outside domain because these particles and their carrying energies are not counted.

![Graph (a): Numerical energy.](image)

![Graph (b): Particle number.](image)

**Fig. 4.** Numerical energy and particle number under PAUM: The particle number after resampling increases from $2.38 \times 10^9$ to $1.45 \times 10^{10}$, while the total mass decreases to $5.74 \times 10^9$. Numerical energy increases from $-0.319 \text{ a.u.}$ to $-0.157 \text{ a.u.}$, partially caused by that the total mass outside the domain is not counted.

**Particle number:** We are interested in the relation between the growth of particles and the partition level $K$. Fig. 4(b) compares the particle number after PAUM $P + M$, the partition level $K$ and the effective particle number $P - M$. It is found that $P + M$ increases rapidly from $2.38 \times 10^9$ to $1.45 \times 10^{10}$ and becomes stable after $t = 40 \text{ a.u.}$, and the peak of total particle number after branching is about $5 \times 10^{10}$ (here $K \approx 1.1 \times 10^{11}$). The counted total mass decrease from $1 \times 10^9$ to $5.74 \times 10^9$. The growth of particle is significantly suppressed compared with the rate $e^{1.16t}$ without PAUM. This manifests that PAUM is capable of alleviating the sign problem in 6-D phase space.

5.2. **Performance of SPADE.** Now we begin to replace the PAUM by SPADE and adopt the same parameters in the particle generation except the initial effective particle number $N_\alpha = 1 \times 10^7$, so that the initial total particle number $P + M = 2.38 \times 10^7$.

**Performance metrics:** The relative $L^2$-error is adopted to study the accuracy. $f_{\text{PAUM}}(x, k, t)$ and $f_{\text{SPADE}}(x, k, t)$ denote the numerical solutions of the reduced Wigner function (re) obtained by PAUM and SPADE, respectively, with $f_{\text{re}}$ given by

$$f_{\text{re}}(x_1, k_1) = \iiint_{\mathbb{R}^4} f(x_1, x_2, x_3, k_1, k_2, k_3) dx_2 dx_3 dk_2 dk_3,$$

This can be approximated by a piecewise function

$$f_{\text{re}}(x_1, k_1) \approx \sum_{i=1}^{60} \sum_{j=1}^{80} \frac{\langle 1_{\mathcal{X}_i \times \mathcal{K}_j}, \mu \rangle \cdot 1_{\mathcal{X}_i \times \mathcal{K}_j}(x, k)}{\text{vol}(\mathcal{X}_i) \text{vol}(\mathcal{K}_j)},$$

where $\mu$ is empirical signed measure in the estimator (2.7).

The relative errors are written as

$$\text{err}_{\text{wfd}}(t) = \frac{\int_{\mathcal{X} \times \mathcal{K}} (\Delta f(x, k, t))^2 dxdk}{\int_{\mathcal{X} \times \mathcal{K}} (f_{\text{PAUM}}(x, k, t))^2 dxdk}.$$
where $\Delta f(x, k, t) = |f^\text{SPADE}(x, k, t) - f^\text{PAUM}(x, k, t)|$, and the integrals above are evaluated using Eq. (5.4) and a simple rectangular rule over the $60 \times 80$ uniform mesh. Meanwhile, we measure corresponding relative errors for physical quantities, e.g., the spatial marginal (sm) probability distribution and the momental marginal distribution (mm) in a similar way, denoted by $\text{err}_{\text{sm}}(t)$ and $\text{err}_{\text{mm}}(t)$.

To obtain a more complete view of the accuracy, we also measure the deviation of the total energy.

**Parameters for SPADE:** For the sake of comparison, we still use the computation domain $X \times K = [-9, 9]^3 \times [-2.4, 2.4]^3$ and no specified boundary condition is adopted as before. We also set the final time $t_{\text{fin}} = 100$ a.u. and perform SPADE every 1 a.u., where the maximal gap is adopted in the decision of binary splitting. For the parameters of the TA-improved algorithm, we set the number of iterations $I = 64$ and the number of independent trials $N_t = 1$. According to our tests, under these parameters can TA-improved algorithm provide a good approximation to the star discrepancy.

**Parallelization:** We use 128 threads in the simulations. The data are distributed into 4 nodes and each node provides 32 cores. All the data in different nodes are transferred via the Message Passing Interference (MPI) standard. Only distributed computing is taken into consideration and OpenMP library is NOT used in those experiments. We adopt a domain decomposition strategy by a binary partition of $X \times K$ to make the particle number in each thread as balanced as possible.

**Snapshots:** Snapshots of the Wigner function at instants $t = 5, 10, 20, 50, 100$ a.u. under different resampling strategies are compared in Fig. 5. The main features of the Wigner function can be captured by SPADE even under a relatively small sample size $N_\alpha = 1 \times 10^7$. Refining the clustering by taking a smaller $\vartheta = 0.005$, the resolution of the Wigner function can be further improved, especially for the region where the Wigner function takes negative values.

**Numerical accuracy:** The relative $L^2$-errors are plotted in Fig. (6) under five choices of parameter $\vartheta$ in sequential clustering. Choosing a small $\vartheta$ refines the adaptive partition, and diminishes the numerical errors. One can see that when $\vartheta = 0.005$ the relative $L^2$-errors do not exceed 35% even up to 100 a.u., which manifests the feasibility of SPADE in long-time simulations.

**Energy conservation:** Larger deviations of numerical energy in SPADE are observed in Fig. 6(d) compared with that in PAUM, but can be systematically suppressed by refining the adaptive partition. The loss of energy is in part induced by that the mass outside the domain is not counted, as shown in Fig. 6(f).

**Particle number:** An interesting observation is that the total particle number $P + M$ will not increase in SPADE as shown in Fig. 6(e), whereas that in PAUM will grow dramatically. This demonstrates the most important feature of SPADE, that is, the partition level $K$ is usually smaller than the particle number $P + M$. In this occasion, it may get rid of the overfitting problem. By contrast, the PAUM requires the sample size to match the fixed partition level, otherwise particles will not be annihilated efficiently. For long-time simulations, the effective particle number $P - M$ seems to decrease faster in SPADE compared with PAUM, indicating that more particles move outside the domain.
Fig. 5. A comparison between PAUM and SPADE (left: PAUM, mid: SPADE, $\theta = 0.005$, right: SPADE, $\theta = 0.02$): The main features of the Wigner function can be captured by SPADE under a small sample size $N_\alpha = 1 \times 10^7$. Numerical resolution can be improved by refining the partition, especially the region where the Wigner function takes negative values.
5.3. Transitions in energy levels. A direct application of the Wigner dynamics is to investigate the transition of energy levels of a quantum system via the spectrum analysis of time series of signals. One can refer to\(^2\) for more details, in which several typical quantum systems in 2-D phase space are studied. We would like to demonstrate that the WBRW-SPA combined with SPADE is able to obtain several important quantum observables at a significant reduction in computational cost from 20 days to 13 hours.

The time series \(\langle x_1 \rangle\) and \(\langle k_1 \rangle\) from \(t = 0\) a.u. to \(t = 100\) a.u. and the corresponding power spectrums are plotted in Fig. 7. Under different annihilation strategies, an oscillating pattern of signals is clearly seen. By analyzing the power spectrum, we obtain several peaks as collected in Table 1. The peak 0.3770 corresponding to the
transition from 1s state to 2s state is observed in all experiments (a slight deviation is observed only for SPADE under $\vartheta = 0.01$). Since the Gaussian Wigner function is essentially a mixed state and may occupy other states, it can probably capture the excitation of electron to higher energy levels.

When PAUM is adopted, we obtain five peaks: 0.0628, 0.2513, 0.3770, 0.5027, 0.8796 and three of them have a correspondence to transitions between the energy level pairs (1, 2), (2, 3) and (1, $\infty$), while two (0.2513, 0.8796) are artificial.

When SPADE is adopted, the artificial peak 0.8796 disappears. Apart from the above energy level pairs, two additional peaks 0.1250 and 0.4444 can also be accurately captured, corresponding to the transitions between the energy level pairs (2, $\infty$) and (1, 3), respectively, while an artificial peak 0.6912 is found under $\vartheta = 0.01$. In practice, artificial peaks can be excluded by multiple trials under different choices of $\vartheta$.

Fig. 7. Time series of signals (left) and their power spectrums (right): Several peaks in the spectrum have a correspondence to the differences in Hydrogen energy levels.

5.4. Benchmark of SPADE: Sample size $N_\alpha$. We fix the parameter $\vartheta$ in adaptive clustering and evaluate the performance of SPADE under various choices of particle number $N_\alpha$. Two groups of simulations are performed under $\vartheta = 0.02$ and $\vartheta = 0.04$, and the choices of $N_\alpha$ include $4 \times 10^5$, $1 \times 10^6$, $1 \times 10^7$ and $1 \times 10^8$. For the testing purpose, we only use 64 threads in SPADE and the final time is set to be $t_{fin} = 10$ a.u.

Fig. 8 plots the time evolution of relative errors of $\text{err}_{wf}$, $\text{err}_{sm}$ and $\text{err}_{mm}$. Fig. 9 records the partition level $K$ in two groups of simulations. The growth of particles and the time evolution of the numerical energy are presented in Fig. 10. Based on these numerical results, we are able to figure out the following observations.

(1) **Convergence with respect to $N_\alpha$**: According to Fig. 8, the numerical accuracy
Energy level transitions with initial 1s-2s superposition state: $\Delta E_{mn}$ denotes the energy difference for the transition between the $n$-th and the $m$-th levels. The reference (Ref.) value of $\Delta E_{mn}$ is calculated by $\Delta E_{mn} = |E_m - E_n|$ whereas the numerical (Num.) value is directly obtained by the spectral analysis of either the averaged displacement $\langle x_1 \rangle$ or momentum $\langle k_1 \rangle$ from the numerical evolution of the Wigner equation until $t = 100$ a.u.

| Annihilation | $m$ | $n$ | $E_m$ | $E_n$ | $\Delta E_{mn}$ | Num. Error | Rel. |
|--------------|-----|-----|-------|-------|-----------------|------------|-----|
| PAUM         | 1   | 2   | -0.5000 | -0.1250 | 0.3750 | 0.0020 | 0.53% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.0027 | 0.54% |
| $\vartheta = 0.005$ | 2   | 3 | -0.1250 | -0.0556 | 0.0694 | 0.0628 | 0.0066 | 10.51% |
|              | 1   | 2 | -0.5000 | -0.1250 | 0.3750 | 0.3142 | 0.0608 | 16.21% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.5027 | 0.0027 | 0.54% |
| $\vartheta = 0.01$ | 2   | 3 | -0.1250 | -0.0556 | 0.0694 | 0.0628 | 0.0066 | 10.51% |
|              | 1   | 2 | -0.5000 | -0.1250 | 0.3750 | 0.3142 | 0.0608 | 16.21% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.5027 | 0.0027 | 0.54% |
| $\vartheta = 0.02$ | 2   | 3 | -0.1250 | -0.0556 | 0.0694 | 0.0628 | 0.0066 | 10.51% |
|              | 1   | 2 | -0.5000 | -0.1250 | 0.3750 | 0.3142 | 0.0608 | 16.21% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.5027 | 0.0027 | 0.54% |
| $\vartheta = 0.04$ | 2   | 3 | -0.1250 | -0.0556 | 0.0694 | 0.0628 | 0.0066 | 10.51% |
|              | 1   | 2 | -0.5000 | -0.1250 | 0.3750 | 0.3142 | 0.0608 | 16.21% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.5027 | 0.0027 | 0.54% |
| $\vartheta = 0.08$ | 2   | 3 | -0.1250 | -0.0556 | 0.0694 | 0.0628 | 0.0066 | 10.51% |
|              | 1   | 2 | -0.5000 | -0.1250 | 0.3750 | 0.3142 | 0.0608 | 16.21% |
|              | 1   | $\infty$ | -0.5000 | 0.0000 | 0.5000 | 0.5027 | 0.0027 | 0.54% |

Table 1

can be improved by increasing $N_\alpha$ from $4 \times 10^5$ to $1 \times 10^8$. The solutions under $\vartheta = 0.02$ are better than those under $\vartheta = 0.04$, indicating that the accuracy is also influenced by the partition level of adaptive clustering.

(2) **Partition level $K$:** According to Figs. 9(b) and 9(c), there is an oscillating pattern in $K$. It actually coincides with the oscillations of $L^2$-errors observed in Fig. 8. Thus the numerical accuracy is closely related to the partition level $K$. Despite some fluctuations, $K$ almost remains at the same level in time evolution. Under a smaller $\vartheta$, the partition is refined, resulting in a larger $K$. The relation between $N_\alpha$ and $K$ is presented in Fig. 9(a). One can observe that $K$ is not linearly dependent on $N_\alpha$, but seems to be $K \propto N_\alpha^{0.3}$.

(3) **Particle growth:** $(P + M)/N_\alpha$ before and after annihilation at instant $t$ are depicted in Fig. 10(a) by dash and solid lines, respectively. The total particle number grows in stochastic simulations and sharply decreases after annihilation. It clearly presents the bottom line structure as observed in 2-D grid-based annihilation, which describes the minimal amount of particles that can accurately capture the oscillating nature of the Wigner function. The
Fig. 8. Convergence with respect to $N_\alpha$ in SPADE: The numerical accuracy can be systematically improved by increasing $N_\alpha$ from $4 \times 10^5$ to $1 \times 10^8$.

bottom line seems to be independent of the choice of $N_\alpha$. Therefore, SPADE can achieve an efficient annihilation by automatically finding an appropriate partition level and consequently alleviate the “overfitting” phenomenon.

(4) **Numerical energy:** Numerical energies in two groups of simulations are plotted in Fig. 10(b), where only 0.4% of total mass is not counted at $t = 10\text{a.u.}$ One can observe that the fluctuation of energy is suppressed when $N_\alpha$ increases, which coincides with the results in Fig. 8. Actually, a suitable setting of parameters in simulations can be determined by just monitoring and diminishing the fluctuation of the numerical energy.

(5) **Computational time:** We record the total wall time for sequential clustering, which occupies more than 85% of total computational time, and $P + M$ at $t_{\text{fin}} = 10\text{a.u.}$ in Table 2. It seems that the computational time seems to
The relation between $N_\alpha$ and $K$.

Fig. 9. The relation between sample size $N_\alpha$ and partition level $K$: Despite some fluctuations, $K$ almost remains at the same level in time evolution. With smaller $\vartheta$, the partition is further refined, resulting in a larger $K$ and $K \propto N_\alpha^{0.3}$

Table 2

| $\vartheta$ | $N_\alpha$ | $P + M$ | time(h) |
|------------|------------|--------|--------|
| 0.02       | $1 \times 10^6$ | $1.26 \times 10^6$ | 11.410 |
|            | $1 \times 10^7$ | $1.27 \times 10^7$ | 74.2085 |
|            | $1 \times 10^8$ | $1.36 \times 10^8$ | 895.097 |
| 0.04       | $1 \times 10^6$ | $1.08 \times 10^6$ | 6.915  |
|            | $1 \times 10^7$ | $1.18 \times 10^7$ | 52.466 |
|            | $1 \times 10^8$ | $1.28 \times 10^8$ | 612.911 |

5.5. Benchmark of SPADE: Partition level $K$ and the parameter $\vartheta$.

Now we turn to investigate how the parameter $\vartheta$ influences the partition level $K$ and numerical accuracy. Fig. 11 plots the time evolution of relative errors of err_{wf}, err_{sm} and err_{mm}. Fig. 12 records the size of partition $K$ in two groups of simulations. The particle growth and numerical energy are presented in Fig. 13. Based on these numerical results, we are able to find out the following observations.

(1) Convergence with respect to $\vartheta$: According to Fig. 11, the numerical accuracy can be improved by decreasing $\vartheta$ from 0.16 to 0.01, because of the refinement in adaptive partition. Also a fluctuation of numerical errors is observed under
Fig. 10. Particle number and numerical energy: SPADE achieves an efficient annihilation of particles and a bottom line of $P + M$, that seems to be independent of the choice of $N_\alpha$, is clearly observed. A small deviation of energy can be achieved when $N_\alpha = 1 \times 10^7$ and $N_\alpha = 1 \times 10^8$, where only 0.4% of total mass is not counted at $t = 10$ a.u.

different $\vartheta$, corresponding to varying partition levels in SPADE. We can see that the solutions under $N_\alpha = 1 \times 10^7$ are much better than those under $N_\alpha = 1 \times 10^6$, which again verifies the convergence with respect to $N_\alpha$.

(2) **Partition level** $K$: It is clearly seen that $K$ increases along with a smaller $\vartheta$, and they seem to be linearly dependent. This gives us a hint to postulate the partition level $K$ under a very small $\vartheta$ before performing the real simulations.

(3) **Particle growth**: $(P + M)/N_\alpha$ before and after annihilation are also depicted in Fig. 13(a) by dash and solid lines, respectively, and the bottom line structure is also observed. When $N_\alpha$ is sufficiently large, the bottom line is independent of the choice of $\vartheta$ as well as the partition level $K$. A special case is that both $N_\alpha$ and $\vartheta$ are small, in which the bottom line moves up to ensure the accuracy.

(4) **Numerical energy**: Numerical energies in two groups of simulations are plotted in Fig. 13, where only 0.4% of total mass is not counted at $t = 10$ a.u. One can observe that the deviations of energy are significantly suppressed by a refinement of the adaptive partition under a smaller $\vartheta$.

(5) **Computational time**: We also record the total wall time for sequential clustering in these experiments and $P + M$ after annihilation at $t_{fin} = 10$ a.u. in Table 3. It is readily seen there that the computational time significantly increases when a smaller $\vartheta$ is chosen. Under different settings of $\vartheta$, $P + M$ remains at the same level of $N_\alpha$. This coincides with the bottom line structure observed in Fig. 13(a).
Fig. 11. Convergence with respect to $\vartheta$ in SPADE: The numerical accuracy can be improved by reducing $\vartheta = 0.16$ to $\vartheta = 0.01$, resulting in a refinement in adaptive partition.

Table 3

| $\vartheta$ | $N_\alpha = 1 \times 10^6$ | $N_\alpha = 1 \times 10^7$ |
|-------------|---------------------------|---------------------------|
| $P + M$     |                           |                           |
| 0.01        | $1.81 \times 10^6$        | $1.41 \times 10^7$        |
| 0.04        | $1.08 \times 10^6$        | $1.18 \times 10^7$        |
| 0.16        | $0.98 \times 10^6$        | $1.01 \times 10^7$        |
| time(h)     | 23.579                    | 125.975                   |
|             | 6.915                     | 52.466                    |
|             | 3.700                     | 35.289                    |

6. Conclusion and discussion. This paper discusses the particle annihilation algorithms to overcome the numerical sign problem in 6-D stochastic Wigner sim-
Fig. 12. The relation between the parameter $\vartheta$ and partition level $K$: Despite some fluctuations, $K$ almost remains at the same level in time evolution. With smaller $\vartheta$, the partition is further refined, resulting in a larger $K$ and $K \propto \vartheta^{-1}$.

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Fig. 13. Particle number and numerical energy: SPADE achieves an efficient annihilation of particles and a bottom line of $P + M$, that seems to be independent of the choice of $\vartheta$, is clearly observed. A small deviation of energy can be achieved when $N_\alpha = 1 \times 10^7$ and $\vartheta = 0.01$, where only 0.4% of total mass is not counted at $t = 10 \text{a.u.}$.

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Appendix A. Gaussian expansion of Hydrogen Wigner function. The Wigner function of the fundamental quantum systems like the hydrogen atom may be useful for quantum tomography, quantum state diagnostic and phase space visualization of negative structures of quantum interference. In practice, the Hydrogen Wigner function can be approximated by the linear combination of GWFs.

The coefficients $a_j$ and $c_j$ of GTOs of the hydrogen atom can be determined by minimizing the energy integral under the potential $V(x)$ in atomic unit (a.u.) $m = \hbar = e = 1$.

\begin{equation}
E[\phi] = \int_{\mathbb{R}^3} \phi^*(x) \left( -\frac{1}{2} \Delta - \frac{1}{|x - x_A|} \right) \phi(x) dx.
\end{equation}

The coefficients of GTOs under different $N_{\text{basis}}$ are collected in Table 4.35
Table 4

Coefficients in the Gaussian-type orbitals for the hydrogen atom.

| Orbit | \(N_{basis}\) | \(a_1\) | \(b_1\) | \(a_2\) | \(b_2\) | \(a_3\) | \(b_3\) | \(a_4\) | \(b_4\) | \(R\) | \(N\) | Energy (a.u.) |
|-------|-------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|--------------|
| 1s    | 1           | 0.28294 | 1.0   | -     | -     | -     | 0.0   | 1.0   | -0.424412  | 1.3414  | 1.0  | 0.20121  |
|       | 2           | 1.3414 | 1.0   | 0.20121 | 3.0 | -     | -     | 0.0   | 3.64921    | -0.45689 | 3.0  | -0.49079  |
|       | 3           | 4.4511 | 1.0   | 0.6766 | 5.721 | 0.1509 | 9.03  | 0.0   | 13.9858    | -0.496979 | 4.4511 | 1.0  | 0.6766   |
|       | 4           | 2.83904 | 1.0   | 0.578897 | 5.90 | 0.139552 | 6.70  | 0.156 | 11.26155 | -0.496979 | 2.83904 | 1.0  | 0.578897 |
| 2s    | 1           | 1.4    | 1.0   | 0.032  | 13.0 | 0.016 | 19.0  | 0.0   | 27.776    | -0.12444 | 1.4  | 1.0  | 0.032    |
|       | 2           | 0.16875 | 1.0   | 0.03375 | 6.1483 | -     | -     | 0.0   | 2.16435    | -0.1230813 | 0.16875 | 1.0  | 0.03375  |
|       | 3           | 0.096649 | 1.0   | 0.073356 | 9.0 | 0.0244519 | 17.3  | 0.0   | 3.59558    | -0.124692 | 0.096649 | 1.0  | 0.073356 |
| 2p_x  | 1           | 0.035943 | 1.0   | 0.035943 | 6.04 | -     | -     | 0.0   | 8.28414    | -0.123684 | 0.035943 | 1.0  | 0.035943 |
|       | 2           | 0.040389 | 1.0   | -     | -     | -     | -     | 0.0   | 3.94264    | -0.118436 | 0.040389 | 1.0  | -     |
|       | 3           | 0.039649 | 1.0   | 0.073356 | 9.0 | 0.0244519 | 17.3  | 0.0   | 3.59558    | -0.124692 | 0.039649 | 1.0  | 0.073356 |

The principal quantum number \(n = 1\) corresponds to the 1s orbital with exact energy \(-0.5\) a.u.,

\[
\phi_{1s}(x) = \frac{1}{\sqrt{\pi}} \exp(-|x|) \approx \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(x).
\]

The principal quantum number \(n = 2\) and the azimuthal quantum number \(l = 0\) corresponds to 2s orbital with exact energy \(-0.125\) a.u.,

\[
\phi_{2s}(x) = \left( \frac{1}{32\pi} \right)^{1/2} (2 - |x|) \exp(-|x|/2) \approx \frac{1}{N} \sum_{j=1}^{3} c_j \phi_j(x) - 1.3159 \phi_{1s}(x)
\]

where \(\phi_{1s}\) is given by Eq. (A.2) for \(N_{basis} = 4\) without normalization.

The characterization of 2p orbital, with principal quantum number \(n = 2\) and the azimuthal quantum number \(l = 1\), becomes a little more complicated as the orbital is no longer spherically symmetry. Under the spherical coordinate, the 2p orbital with exact energy \(-0.125\) a.u. reads that

\[
\phi_{2p_x} = \left( \frac{1}{32\pi} \right)^{1/2} r \cos \vartheta \exp(-r/2) \approx \phi_+(x) - \phi_-(x).
\]

with two lobe Gaussian functions \(\phi_\pm(x)\) to characterize the polarization of orbitals

\[
\phi_+(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x - R|), \quad \phi_-(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x + R|),
\]

and centers \(R = (\pm R, 0, 0)\).

The 3d_{xy} orbital, with the exact energy is \(-0.0555\) a.u., can be approximated by four lobe Gaussian functions,

\[
\phi_{3d_{xy}} \approx \phi_{++}(x) - \phi_{+-}(x) + \phi_{-+}(x) - \phi_{--}(x),
\]

where

\[
\phi_{++}(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x - R_x|), \quad \phi_{+-}(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x + R_x|),
\]

\[
\phi_{-+}(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x - R_y|), \quad \phi_{--}(x) = \frac{1}{N} \sum_{j=1}^{N_{basis}} c_j \phi_j(|x + R_y|),
\]
with \( R_x = (R/2, 0, 0) \) and \( R_y = (0, R/2, 0) \).

A visualization of the spatial distribution (sm), the reduced Wigner function (re) and the marginal distribution (mm) for the hydrogen atom is given in Fig. 14. It is seen that the 1s and 2s states are spherically symmetric in their spatial distributions, while the 2px and 3dxy states are polarized and forbidden at the centre \((x_1, x_2) = (0, 0)\). The spatial distribution spreads into a wider range as the principal quantum number \( n \) increases.

The Wigner function also provides a convenient way to visualize the momental density. Here we plot its projection onto the \((k_1-k_2)\) plane. The momentum distribution is much more narrow than the spatial density since they constitute a pair of Fourier conjugates. It become more and more localized as the principal quantum number \( n \) increases. We realize that 1s and 2s states are spherically symmetric in momental distribution, while the 2px and 3dxy states in momental space are similarly polarized and forbidden at the centre \((k_1, k_2) = (0, 0)\).

The reduced Wigner function is visualized by a projection onto the \((x_1-k_1)\) plane. One can realize that the Wigner functions of 1s and 2s states have positive peaks at \((x_1, k_1) = (0, 0)\), whereas those of 2p and 3d states have negative valleys at the centre. The oscillatory structure of the Wigner function in the phase space is clearly observed. This coincides with our knowledge because negative probability indicates a forbidden region due to the Heisenberg uncertainty principle. In addition, for the states with larger principal quantum number, the Wigner function tends to be more localized in momental space and spreads more widely in spatial space.
Fig. 14. The hydrogen atom: Visualization of the spatial distribution, the Wigner function and the momental distribution. The negative value of the Wigner function presents the forbidden region due to the Heisenberg uncertainty principle.