High-order Path Integral Monte Carlo methods for solving quantum dot problems

Siu A. Chin

Department of Physics and Astronomy,
Texas A&M University, College Station, TX 77843, USA

Abstract

The conventional second-order Path Integral Monte Carlo method is plagued with the sign problem in solving many-fermion systems. This is due to the large number of anti-symmetric free fermion propagators that are needed to extract the ground state wave function at large imaginary time. In this work, we show that optimized fourth-order Path Integral Monte Carlo methods, which use no more than 5 free-fermion propagators, can yield accurate quantum dot energies for up to 20 polarized electrons with the use of the Hamiltonian energy estimator.
The Path Integral Monte Carlo (PIMC) method remains problematic for solving many-fermion systems due to the “sign” problem. Since the anti-symmetric free-fermion propagator (FFP) is not positive-definite, only its magnitude can be sampled by the Monte Carlo method and observables must then be weighted by the overall sign of all FFP in the path-integral. While a suggested remedy has been proposed\textsuperscript{1–3}, the most practical solution in traditional PIMC is to simply ignore the sign-problem by invoking some “fixed-node” or “restricted-path” approximations\textsuperscript{4}. Here, we propose a new way of alleviating the sign problem by drastically reducing the number FFP needed in PIMC calculations.

In conventional PIMC, the second-order primitive action (PA) propagator with one FFP is only accurate at very small time steps. Thus hundreds of PA propagators are needed to extract the ground state at large imaginary times. When one has hundreds of FFP, then whether one is sampling the permutations\textsuperscript{4} or directly evaluating the anti-symmetric free-propagator\textsuperscript{5}, the average of the sign is close to zero and the sign-problem is intractable. However, the severity of the sign problem (only occurs in more than one dimension\textsuperscript{5}) depends critically on the number free-fermion propagators used. Since only the trace of the density matrix is computed in PIMC, the trace of one or two FFP is always positive, with no sign problem. The average of the overall sign then decreases rapidly toward zero with increasing numbers of FFP and fermions. Thus if the number of FFP can be drastically reduced, say to less than 10, then one may be able to extract ground state properties before the sign problem gets severe. This is the key idea of this work.

This idea has not been contemplated before, because it has always been deemed impossible to reach the ground state with so few FFP using the PA propagator. However, in the bosonic case of liquid helium, we have shown\textsuperscript{6} that even a single use of a fourth-order propagator can reach closer to the ground state than many elaborate variational schemes. Recently, the ground state energy of liquid Helium has been computed in PIMC by Sakkos, Casulleras and Boronat\textsuperscript{6}, using a fourth-order propagator that can be “fine-tuned” to converge at the sixth-order for the energy. Their work reduces, by an order-of-magnitude, the number of propagators (or beads) needed in bosonic PIMC. Subsequently, R. Rota et al.\textsuperscript{8} showed that a Ground-State PIMC (GSPIMC) calculation only needed about ten propagators to achieve the same objective! Similar results have also been obtained for the same system with GSPIMC by Zillich, Mayrhofer and Chin\textsuperscript{9}, using sixth and eighth-order extrapolated propagators. The tremendous success of these results have inspired this work to apply
higher-order PIMC methods to fermions in quantum dots.

Aside from using higher-order propagators, these calculations also have in common in computing the energy from the Hamiltonian. By the Golden-Thompson inequality, the thermodynamics estimator used in conventional PIMC converges to the ground state energy only from below. In the few-propagators case, it is so far from convergence that it is totally useless. The use of the virial estimator is risky for quantum dots, since it may dip below the exact ground state energy. In this work, we follow the success of the GSPIMC method in also using the Hamiltonian estimator in PIMC. This estimator is known in Bosonic PIMC, but we generalize it here to include the anti-symmetric, determinant propagator. There are three advantages in using the Hamiltonian estimator in PIMC. First, it gives a variational upper-bound to the ground state energy, as in GSPIMC. Second, its result can be double-checked by use of a variant, the Clark-Westhaus (CW) form of the kinetic energy. Third, in contrast to GSPIMC, no trial ground-state wave function is needed.

The Hamiltonian for $N$ electrons in a 2D harmonic dot is

$$H = \sum_{i=1}^{N} \left( -\frac{\hbar^2}{2m^*} \nabla_i^2 + \frac{1}{2} m^* \omega^2 r_i^2 \right) + \sum_{i<j} \frac{e^2}{\kappa |r_i - r_j|}. \tag{1}$$

By expressing $r_i = \ell_0 x_i$, where $\ell_0 = \sqrt{\hbar/(m^* \omega)}$ is the harmonic length, one obtains the dimensionless Hamiltonian $\mathcal{H}$ in terms of dimensionless vectors $x_i$

$$\mathcal{H} \equiv \frac{H}{\hbar \omega} = \sum_{i=1}^{N} \left( -\frac{1}{2} \nabla_i^2 + \frac{1}{2} x_i^2 \right) + \sum_{i<j} \frac{\lambda}{|x_i - x_j|} \tag{2}$$

with effective coupling strength $\lambda = \ell_0 m^* e^2/(\hbar^2 \kappa) = l_0 a_B^{-1}$, where $a_B$ is the effective Bohr radius. The Hamiltonian $\mathcal{H}$ is the quantum dot’s energy in units of $\hbar \omega$.

The corresponding imaginary time propagator is

$$G(\tau) = e^{-\beta H/\hbar} = e^{-\tau \mathcal{H}} = e^{-\tau(T+V)}, \tag{3}$$

where $\tau = \beta \omega$ is the dimensionless imaginary time, and $T$ and $V$ are the kinetic and potential operators of $\mathcal{H}$. The PA propagator approximates $G(\epsilon)$ at small time $\epsilon = \tau/n$ as

$$G_2(\epsilon) = e^{-\epsilon V/2} e^{-\epsilon T} e^{-\epsilon V/2} + O(\epsilon^3), \tag{4}$$

with coordinate representation

$$G_2(\mathbf{X}, \mathbf{X}'; \epsilon) = \langle \mathbf{X} | G_2(\epsilon) | \mathbf{X}' \rangle = e^{-\epsilon V(\mathbf{X})/2} G_0(\mathbf{X}, \mathbf{X}'; \epsilon) e^{-\epsilon V(\mathbf{X}')/2}, \tag{5}$$
where $X = \{x_i\}$ is the position vector of all $N$ fermions, and

$$V(X) = \sum_{i=1}^{N} \frac{1}{2} x_i^2 + \sum_{i<j}^{N} \frac{\lambda}{|x_i - x_j|}. \quad (6)$$

The anti-symmetric FFP $G_0(X, X'; \epsilon)$ is given by

$$G_0(X, X'; \epsilon) = \langle X | e^{-\epsilon T} | X' \rangle = (2\pi\epsilon)^{-ND/2} \text{det} M, \quad (7)$$

where $N$ is the number of fermions (electrons of the same spin), $D$ is the dimension of the system, and $M$ is the $N \times N$ anti-symmetric diffusion matrix

$$M_{ij}(X, X') = \exp \left[ -\frac{1}{2\epsilon} (x_i - x'_j)^2 \right]. \quad (8)$$

For computing the energy, it is convenient to write

$$G_0(X, X'; \epsilon) = e^{-u_0(X, X'; \epsilon)},$$

$$u_0(X, X'; \epsilon) = \frac{ND}{2} \ln(\epsilon) - \ln(\text{det} M). \quad (9)$$

In PIMC, the energy is calculated from ($X_k$ is denoted simply by $k$)

$$E = \int d1...dn G_2(1, 2; \epsilon) \mathcal{H} G_2(2, 3; \epsilon) \cdots G_2(n, 1; \epsilon) \over \int d1...dn G_2(1, 2; \epsilon) G_2(2, 3; \epsilon) \cdots G_2(n, 1; \epsilon) \quad (10)$$

and averaged over all $n$ places where $\mathcal{H}$ can be inserted between propagators. Since the FFP is not positive-definite, the above integral is sampled as

$$E = \frac{\int d1...dn \, \text{sgn} E_{\mathcal{H}}(k, k + 1) P(1, 2, \cdots n; \epsilon)}{\int d1...dn \, \text{sgn} P(1, 2, \cdots n; \epsilon)} \quad (11)$$

with the probability distribution function taken to be

$$P(1, 2, \cdots n; \epsilon) = |G_2(1, 2; \epsilon) G_2(2, 3; \epsilon) \cdots G_2(n, 1; \epsilon)|, \quad (12)$$

and where sgn = ±1 is the overall sign of the product of $G_2$’s. The Hamiltonian energy estimator is given by

$$E_{\mathcal{H}}(X, X'; \epsilon) = \frac{\mathcal{H} G_2(X, X'; \epsilon)}{G_2(X, X'; \epsilon)}$$

$$= \sum_{i=1}^{N} (-\frac{1}{2} \nabla_i^2) G_2(X, X'; \epsilon) G_2(X, X'; \epsilon) + V(X) \quad (13)$$
The alternative CW form of the kinetic energy is to let one of the gradient operator acts to the left, giving

\[
E_{CW}(X^*, X, X'; \epsilon) = \sum_{i=1}^{N} \frac{1}{2} G_2(X^*, X; \epsilon) \nabla_i \cdot \nabla_i G_2(X, X'; \epsilon) + V(X). \tag{14}
\]

The exact (and the free) propagator satisfies the equation

\[
-\frac{\partial}{\partial \epsilon} G(X, X'; \epsilon) = \mathcal{H} G(X, X'; \epsilon). \tag{15}
\]

This equality no longer holds when \(G(X, X'; \epsilon)\) is replaced by an approximation, such as \(G_2(X, X'; \epsilon)\). In this case, when both sides of the equation are divided by \(G_2(X, X'; \epsilon)\), the RHS gives the “Hamiltonian” estimator as stated above. The LHS then gives the “Thermodynamics” energy estimator

\[
E_{TH}(X, X'; \epsilon) = \frac{-\partial_i G_2(X, X'; \epsilon)}{G_2(X, X'; \epsilon)} = \frac{\partial}{\partial \epsilon} u_0(X, X'; \epsilon) + \frac{1}{2} [V(X) + V(X')]. \tag{16}
\]

By the repeated use of the identity

\[
\frac{\partial}{\partial \alpha} \ln(\det M) = \text{Tr} \left[ M^{-1} \frac{\partial M}{\partial \alpha} \right], \tag{17}
\]

all three estimators can be computed without difficulties:

\[
E_{TH}(X, X'; \epsilon) = \frac{ND}{2\epsilon} - \frac{1}{2\epsilon^2} \sum_{i=1}^{N} (x_i^2 + x_i'^2 - 2x_i \cdot \tilde{x}_i') + \frac{1}{2} [V(X) + V(X')], \tag{18}
\]

\[
E_{H}(X, X'; \epsilon) = \frac{1}{2} \sum_{i=1}^{N} \nabla_i^2 \left[ u_0 + \frac{\epsilon}{2} V \right] - \frac{1}{2} \sum_{i=1}^{N} \left[ \nabla_i (u_0 + \frac{\epsilon}{2} V) \right]^2 + V(X), \tag{19}
\]

\[
E_{CW}(X^*, X, X'; \epsilon) = \frac{1}{2} \sum_{i=1}^{N} \nabla_i \left[ u_0(X, X^*; \epsilon) + \frac{\epsilon}{2} V \right] \cdot \nabla_i \left[ u_0(X, X'; \epsilon) + \frac{\epsilon}{2} V \right] + V(X), \tag{20}
\]
where
\[
\nabla_i u_0(X, X'; \epsilon) = \frac{1}{\epsilon} (x_i - \tilde{x}'_i), \\
\nabla^2_i u_0(X, X'; \epsilon) = \frac{D}{\epsilon} - \frac{1}{\epsilon^2} (x'^2_i - \tilde{x}'^2_i). \\
\]
(21)

and where \( \tilde{x}'_i \) is defined by
\[
\tilde{x}'_i \equiv \sum_{k=1}^{N} x'_k M_{ik}(X, X') M^{-1}_{ki}(X, X'). \\
\]
(22)

Thus in all three energy estimates, the calculation of \( M^{-1} \) is required. In the free propagator case, one has indeed \( E_{\text{TH}} = E_{\text{H}} \). The CW estimator will generally have greater variance than the Hamiltonian estimator.

After a set of \( M \) configurations \( \{X^{(m)}_i\} \) has been generated according to \( P(X_1, X_2, \cdots X_n; \epsilon) \), the energy can be computed by using the above three estimators as
\[
E = \frac{\sum_{m=1}^{M} \text{sgn}_k \left[ \frac{1}{n} \sum_{k=1}^{n} E_{\text{H,CW,TH}}(X^{(m)}_k, X^{(m)}_{k+1}; \epsilon) \right]}{\sum_{k=1}^{M} \text{sgn}_k}. \\
\]
(23)

For \( N = 8 \) spin-polarized electrons at the strong-coupling limit of \( \lambda = 8 \), the the energy of these three estimators are compared in Fig.1. This is the largest quantum dot at the strongest coupling considered in Egger et al.’s PIMC calculation and in Rontani et al.’s configuration-interaction study. The thermodynamics estimator showed no convergence for up to 8 PA propagators, whereas the Hamiltonian and CW estimators are in excellent agreement in providing upper-bounds to the ground state energy from 2 to 8 propagators. The sign problem is completely under control in these calculations. The Hamiltonian energy minimum in the 8 propagators case is already close to the result of Egger et al. This strongly suggests that improving the propagator beyond second-order can circumvent the sign problem in these quantum dot calculations.

The short-time propagator can be approximated to any order by a product decomposition,
\[
e^{-\epsilon(T+V)} = \prod_{i=1}^{n} e^{-t_i \epsilon T} e^{-v_i \epsilon V}, \\
\]
(24)

with a suitable set of coefficients \( \{t_i, v_i\} \). However, as first shown by Sheng, Suzuki, Goldman-Kaper, and more recently in a constructive proof by Chin, beyond second order, any factorization of the form (24) must contain some negative coefficients in the set \( \{t_i, v_i\} \) and cannot be used in PIMC. This is because if \( t_i \) were negative, then replacing
\[ \epsilon \to t_i \epsilon \text{ in the free fermion propagator } \] would result in an unbounded function that cannot be normalized as a probability. This simply reflects the fact that diffusion is a time-irreversible process. To have forward fourth-order schemes, with all positive coefficients, one must include the gradient potential

\[ [V, [T, V]] = \sum_{i=1}^{N} |\nabla_i V|^2, \]  

(25)
FIG. 2. Comparing the Hamiltonian energies from optimized fourth-order propagators vs the PA Hamiltonian energies from Fig. 1 for the same $N = 8$ quantum dot. These optimized propagator results are labeled as “Best Bead” (BB), with 3-5 free-fermion propagators.

in the decomposing process$^{10,20}$. These fourth-order schemes have been used successfully in bosonic DMC and PIMC simulations$^{7,8,21,22}$. Here, we will use a more extended family of these forward fourth-order propagators, with arbitrary numbers of free-fermion $T$ operators as described in Refs.$^{18}$ and $^{23}$.

In order to compare with the PA algorithm, we will characterize these algorithms by their number of $T$ operators, or beads. We will consider approximations of the form (24) with $t_1 = 0$ and with left-right symmetric coefficients $v_1 = v_N$, $t_2 = t_N$, etc.,

$$T_{(N-1)B}^{(4)}(\epsilon) = e^{v_1 \epsilon V} e^{t_2 \epsilon T} e^{v_2 \epsilon V} \cdots e^{t_N \epsilon T} e^{v_N \epsilon V}. \quad (26)$$
FIG. 3. Convergence of the Hamiltonian energy for $N = 20$ polarized electrons using the optimized, fourth-order 3- and 4-bead propagators. Errorbars are computed from 200-300 block-average of $5 \times 10^4$ configurations of all 20 electrons.

This will be fourth-order if one chooses $\{t_i\} > 0$ with $\sum_{i=1}^{N} t_i = 1$, fixes $\{v_i\}$ by

$$v_1 = v_N = \frac{1}{2} + \lambda_2(1 - t_2), \quad v_i = -\lambda_2(t_i + t_{i+1}),$$

(27)

where $\lambda_2 = -\phi^{-1}/2, \quad \phi = 1 - \sum_{i=1}^{N} t_i^2$, and divide the required gradient potential term

$$\frac{1}{24}(\frac{1}{\phi} - 1)\epsilon^3[V, [T, V]]$$

left-right symmetrically among all the $v_i\epsilon V$ terms in (26). In order to compute the Hamiltonian energy as simply as in the PA case (with only minor changes from $\epsilon \rightarrow t_i \epsilon$ and $\epsilon \rightarrow v_i \epsilon$), the gradient potential term must not be distributed to the $v_1$ and $v_N$ potential
TABLE I. Comparison of spin-polarized electron ground state energies $E_0/h\omega$ at coupling $\lambda = \sqrt{3}$ for $N = 2$ (with exact energy $E_0/h\omega = 4$) and at $\lambda = 8$ for $N > 2$.

| N  | 8 PA beads | 3 B-beads | 4 B-beads | 5 B-beads | PIMC$^3$ | CI$^{14}$ | DMC$^{24,25}$ |
|----|------------|-----------|-----------|-----------|----------|----------|------------|
| 2  | 4.042(5)   | 4.033(2)  | 4.014(3)  | 4.001(4)  |          |          |            |
| 3  | 15.694(3)  | 15.66(3)  | 15.63(3)  | 15.610(4) | 15.59(1) | 15.595    |            |
| 4  | 27.92(1)   | 27.898(4) | 27.861(8) | 27.82(2)  | 27.823(11) | 27.828    |            |
| 5  | 43.08(1)   | 43.020(5) | 43.00(3)  | 42.90(2)  | 42.86(4) | 42.88     |            |
| 6  | 60.725(15) | 60.622(6) | 60.53(3)  | 60.46(2)  | 60.42(2) | 60.80     | 60.3924(2) |
| 7  | 80.81(2)   | 80.714(8) | 80.59(2)  | 80.54(3)  | 80.59(4) | 80.5146(2)|            |
| 8  | 103.53(3)  | 103.42(1) | 103.28(2) | 103.18(3) | 103.26(5)| 103.0464(4)|            |
| 9  | 128.6(1)   | 128.37(1) | 128.23(4) | 128.0(1)  |          |          |            |
| 10 | 155.5(4)   | 155.38(1) | 155.21(6) | 154.9(2)  |          |          |            |
| 12 | 215.79(2)  | 215.4(1)  | 215.2(2)  |          |          |          |            |
| 14 | 284.43(2)  | 284.08(8) | 283.6(4)  |          |          |          |            |
| 16 | 360.53(5)  | 360.0(3)  |          |          |          |          |            |
| 18 | 444.04(4)  | 442.9(4)  |          |          |          |          |            |
| 20 | 534.63(4)  | 534.1(2)  |          |          |          |          |            |

terms. The freedom to choose $\{t_i\}$ and to distribute the the gradient potential terms among the remaining $v_i$ potential terms then allows one to fine-tune the propagator to minimize the energy$^{7,8}$.

In Fig.2 we compare the Hamiltonian energy obtained by these optimized fourth-order propagator with those of the PA propagator in Fig.1. The energy is computed by sampling the trace of only a single fourth-order propagator having 3 to 5 beads. One sees that the optimized 3-bead propagator already has energy lower than that of 8 PA propagators. The 5-bead propagator has energy lower than that of Egger et al.'s calculation$^3$. Table II compares our results to those obtained by the PIMC method of Egger et al.$^3$, the configuration-interaction method of Rontani et al.$^{14}$ and the Diffusion Monte Carlo (DMC) method of F. Pederiva et al.$^{24}$ and Ghosal et al.$^{25}$, for up to $N = 20$ spin-polarized electrons. In Fig.3
we show the convergence of the 3 and 4-bead propagators for solving the case of \( N = 20 \) spin-polarized electrons. This is a good illustration of the sudden appearance of the sign problem, which blew up the 4-bead calculation with a large variance at \( \tau = 7 \). Nevertheless, the Hamiltonian estimator still gives excellent upper-bounds to the energy at \( \tau = 6 \) and \( \tau = 8 \).

In this work, we have shown that optimized fourth-order propagators, in using only 3-5 FFP, together with the use of the Hamiltonian estimator, can effectively limit the severity of the sign problem and allow accurate calculation of quantum dot energies for up 20 fermions.

This publication was made possible by NPRP GRANT #5-674-1-114 from the Qatar National Research Fund (a member of Qatar Foundation). The statements made herein are solely the responsibility of the author.

---

1. C. H. Mak, R. Egger, and H. Weber-Gottschick, Phys. Rev. Lett. 81, 4533 (1998).
2. R. Egger, L. Mühlbacher, and C. H. Mak, Phys. Rev. E 61, 5961 (2000).
3. R. Egger, W. Häusler, C. H. Mak and H. Grabert, Phys. Rev. Lett. 82, 3320 (1999); 83, 462(E) (1999).
4. D. M. Ceperley, Rev. Mod. Phys., 67, 279 (1995).
5. M. Takahashi and M. Imada, J. Phys. Soc. Jpn 53, 3765 (1984).
6. Orion Ciftja and Siu A. Chin, Phys. Rev. B 68, 134510 (2003)
7. K. Sakkos, J. Casulleras, and J. Boronat, J. Chem. Phys. 130, 204109 (2009)
8. R. Rota, J. Casulleras, F. Mazzanti, and J. Boronat, Phys. Rev. E 81, 016707 (2010)
9. R. E. Zillich, J. M. Mayrhofer and Siu A. Chin, J. Chem. Phys. 132, 044103 (2010).
10. S. Golden, Phys. Rev. 127, B1127, (1965).
11. C. J. Thompson, J. Math. Phys. 6, 1812 (1965).
12. T. W. Whitfield and G. L. Martyna, J. Chem. Phys. 132, 044103 (2010).
13. J. W. Clark, in Progress in Particle and Nuclear Physics, Vol. II, edited by D. H. Wilkinson (Pergamon, Oxford, 1979), and references cited therein.
14. M. Rontani, C. Cavazzoni, D. Bellucci and G. Goldoni J. Chem. Phys. 124, 124102 (2006).
15. Q. Sheng, IMA J. Num. Anaysis, 9, 199 (1989).
16 M. Suzuki, J. Math. Phys. 32, 400 (1991).
17 D. Goldman and T. J. Kaper, SIAM J. Numer. Anal., 33, 349 (1996).
18 S. A. Chin, Phys. Lett. A 354, 373 (2006)
19 M. Suzuki, *Computer Simulation Studies in Condensed Matter Physics VIII*, eds, D. Landau, K. Mon and H. Shuttler (Springler, Berlin, 1996).
20 S. A. Chin, Physics Letters A 226, 344 (1997).
21 H. A. Forbert and S. A. Chin, Phys. Rev. E 63, 016703 (2001); Phys. Rev. B 63, 144518 (2001).
22 S. Jang, S. Jang and G. A. Voth, J. Chem. Phys. 115 7832, (2001).
23 S. A. Chin, Phys. Rev. E, 73, 026705 (2006).
24 F. Pederiva, C. J. Umrigar and E. Lipparini, Phys. Rev. B 62, 8120 (2000), B 68, 089901(E) (2003)
25 A. Ghosal, A. D. Güclü, C. J. Umrigar, D. Ullmo and H. U. Baranger Phys. Rev. B 76, 085341 (2007)