High-accuracy Trotter-formula method for path integrals

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Path integrals are a powerful method for calculating real time, finite temperature, and ground state properties of quantum systems. By exploiting some remarkable properties of the symmetric Trotter formula and the discrete Fourier transform, we arrive at a high-accuracy method for removing "time slice" errors in Trotter-approximated propagators. We provide an explicit demonstration of the method applied to the two-body density matrix of ^{4}He. Our method is simultaneously fast, high precision, and computationally simple and can be applied to a wide variety of quantum propagators.

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Recent years have seen large scale computational methods that employ a "propagator" such as $\exp(-i\frac{Ht}{\hbar})$ or $\exp(-\frac{H}{k_BT})$ to carry out quantum time evolution [1,2] or calculations in quantum statistical mechanics [3,4]. This paper deals with analysis and enhancement of such propagator schemes when they are based on approximations like the Trotter formula [5],

$$e^{-\tau H} = \lim_{N\to\infty}\left[e^{-\tau T/N}e^{-\tau V/N}\right]^N. \tag{1}$$

(T is the kinetic energy and V the potential energy operator. The "time" variable τ is either $1/k_BT$ or $it/\hbar$.) We will show that a wide class of product approximations have errors proportional to only the *even* powers of $1/N$ and use this property with a Romberg-type integration scheme to successively eliminate time-slice errors to very high order. This approach permits substantial reduction of N, extending the variety of problems that are computationally accessible.

For numerical applications of Eq. (1) with finite N, errors depending on the time slice, $\Delta\tau = \tau/N$, enter through the break up,

$$e^{-\Delta\tau H} \approx e^{-\Delta\tau T}e^{-\Delta\tau V}, \tag{2}$$

which is in error at the level of $(\Delta\tau)^2$. The error accumulated in the N factors in the entire propagator is of order $1/N$. A superior break up was given by Feynman [6] who symmetrized the incremental propagator,

$$e^{-\Delta\tau H} \approx e^{-\frac{\Delta\tau}{2}V}e^{-\Delta\tau T}e^{-\frac{\Delta\tau}{2}V} \equiv U_S(\Delta\tau). \tag{3}$$

The error of the symmetrized propagator, U_S, is order $(\Delta\tau)^3$. This yields a $1/N^2$ error in a Green function or density matrix approximated by successive products of the form

$$\begin{aligned} e^{-\tau H} &\approx U_S(\tau/N)^N \\ &= e^{-\frac{\Delta\tau}{2}V}e^{-\Delta\tau T}e^{-\Delta\tau V}\cdots e^{-\Delta\tau V}e^{-\Delta\tau T}e^{-\frac{\Delta\tau}{2}V}. \end{aligned} \tag{4}$$

Many authors have offered error analyses similar to the above [4,6,7]. However, we found no attempt to exploit the error structure with standard extrapolation methods [8] to eliminate the low order errors resulting from discretization. For example, an operator without the $1/N^2$ error is

$$e^{-\tau H} \approx \left[4U_S(\tau/2N)^{2N} - U_S(\tau/N)^N\right]/3. \tag{5}$$

Implementing this numerically requires up to three times as much computation and double the storage, but the remaining error is of order $1/N^4$. As we show below, this process can be easily generalized so that each time the number of points is doubled, the error is again reduced by a factor of N^2. This enhanced error reduction occurs only when an asymptotic expansion for the error in the break up formula contains only powers of $1/N^2$.

Hatano and Suzuki [9] showed that the break up into N symmetric propagators, i.e., Eq. (4), does not include odd-order powers of $\Delta\tau$. Their proof involved an infinite expansion of commutators. We will show here that this error structure is more general. Our proof is based on the observation that if the propagator formed of N products is an even function of N its error expansion necessarily contains only powers of $1/N^2$. Given any break up, $e^{-\tau H} \approx U(\tau/N)^N$, evenness in N requires that it be invariant under $N \to -N$, i.e.,

$$[U(\tau/N)]^N = [U(-\tau/N)]^{-N}. \tag{6}$$

From this we see that an approximate propagator constructed of N products, each of which satisfies a "unitarity condition,"

$$U(\Delta\tau)U(-\Delta\tau) = 1, \tag{7}$$

will have an asymptotic error expansion with only powers of $1/N^2$.

The simple break up given by Eq. (2) fails to satisfy the unitarity condition. The symmetric propagator, U_S, does satisfy this as do several others, including the exact propagator, $e^{-\Delta\tau H}$, and certain rational approximations such as $(1-\Delta\tau H/2)/(1+\Delta\tau H/2)$. Indeed, if $U(\Delta\tau)$ does not satisfy the unitarity condition, then the construction $U(\Delta\tau/2)U^{-1}(-\Delta\tau/2)$ does.

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To illustrate the successive elimination of error terms in Trotter-approximated propagators, we apply our method to the calculation of the two-body density matrix of helium atoms and employ the symmetric break up of Eq. (3). The real-space two-body density matrix is a function of 12 coordinates. Eliminating the center of mass in the usual fashion leaves a function of six coordinates. The Bloch equation in these coordinates is then

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + v\,(r)\right)\rho(\vec{r},\vec{r}\,',\beta) = -\frac{\partial}{\partial\beta}\rho(\vec{r},\vec{r}\,',\beta), \quad (8)$$

with the boundary condition $\rho(\vec{r},\vec{r}\,',\beta\to 0) = \delta^3(\vec{r}-\vec{r}\,')$. The Hamiltonian operates on the unprimed coordinates, m is the reduced mass and $v(r)$ is the central two-body potential which we take to be the HFDHE2 potential (a potential for He using the Hartree-Fock dispersion method) of Aziz *et al.* [10].

The usual partial wave decomposition can be used to expand in Legendre polynomials of the cosine of the angle between $\vec{r}$ and $\vec{r}\,'$,

$$\rho(\vec{r},\vec{r}\,',\beta) = \sum_{l=0}^{\infty}\frac{(2l+1)\rho_l\,(r,r',\beta)}{4\pi r r'}P_l\,(\hat{r}\cdot\hat{r}\,')\,. \quad (9)$$

The lth partial wave density matrix then satisfies a radial Bloch equation,

$$\left(-\frac{\hbar^2}{2m}\left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2}\right] + v(r)\right)\rho_l(r,r',\beta)$$

$$= -\frac{\partial}{\partial\beta}\rho_l\,(r,r',\beta) \quad (10)$$

with an analogous one-dimensional δ function boundary condition. When we refer to the "potential" for ρ_l below, we will implicitly include the r^{-2} term.

We have solved Eq. (10) for temperatures below 1 K, but we present results at 40 K because this temperature was selected in the bulk ^{4}He calculations of Ceperley and Pollock. They have discussed techniques employed to obtain the accurate two-body density matrix needed in their path integral Monte Carlo calculations [3] and adopt the "matrix squaring" method of Klemm and Storer [11]. We present numerical results for $l = 0$, but our method actually improves for larger l values and we routinely incorporate $l \geq 40$ in our construction of the full two-body density matrix.

Having specified our problem formally, we must develop a discrete representation for numerical solution. Following Eqs. (3) and (4), we break up the propagator for ρ_l into N equal slices of $\beta = 1/k_BT \equiv N\Delta\tau$ and choose a uniform spatial grid, so that the jth radial distance is $j\Delta r$, with $1 < j \leq M-1$, and $M\Delta r$ is large enough that the density matrix is numerically zero at that end point.

Evaluating any short-time propagator at the grid points produces a short-time density matrix,

$$U_{jm} = \langle j|U|m\rangle. \quad (11)$$

The discrete representation of the full density matrix becomes a series of matrix multiples,

$$\rho_l(j_N, j_0, \beta) = \sum_{j_1,j_2,\ldots,j_{N-1}} U_{j_N\,j_{N-1}}\cdots U_{j_2\,j_1}U_{j_1\,j_0}. \quad (12)$$

A small $\Delta\tau = 1/Nk_BT$ increment is equivalent to a large temperature so there are a variety of high temperature, semiclassical approximations that may be employed [3] to obtain the initial matrix in Eq. (11). However, simply evaluating a high temperature density matrix at the spatial grid points will not in general produce a matrix which satisfies the unitarity condition, Eq. (7), even if the original continuous density matrix $\langle r|U|r'\rangle$ does satisfy this condition.

Klemm and Storer [11] developed their "matrix squaring" method based on Eq. (12). If N is a power of 2, then Eq. (12) can be broken up into $\log_2 N$ factors, each one being the square of the previous one. Unless the unitary condition for the matrix U_{jm} is satisfied, the product approximation, Eq. (12), does not have the $1/N^2$ error structure.

A discrete space representation of the symmetric propagator that does satisfy the needed unitary condition can be calculated by Fourier transforms on a discrete basis. Examining Eq. (11) for the symmetric propagator, we see that the potential factors in Eq. (3) are diagonal in the real-space basis, but the kinetic energy operator is not. We insert a plane wave basis on Mdiscrete k points $|k_n\rangle$ [with $k_n r_j = \pi nj/M$ and $\langle j|k_n\rangle = \sin(\pi nj/M)\sqrt{2/M}$]. The matrix element of a single symmetric propagator of Eq. (11) is then

$$U_{jm} = \sum_n \langle j|e^{-\frac{\Delta\tau}{2}V(r_j)}|k_n\rangle \times e^{-\Delta\tau\frac{\hbar^2k_n^2}{2m}}\langle k_n|e^{-\frac{\Delta\tau}{2}V(r_m)}|m\rangle. \quad (13)$$

This form regains the unitarity property, Eq. (7), and the products, Eq. (12), have even-order errors suitable for accurate extrapolation.

Using the unitary U_{jm}, the products for Eq. (12) can be carried out with direct matrix multiplication, the Klemm-Storer matrix squaring technique, or using fast Fourier transform (FFT) methods [2]. Our form of U_{jm} is well suited to the FFT method. This process is initiated with the infinite temperature density matrix $\rho_l(i,j,0) = \delta_{ij}/\Delta r$. Multiply it by $\exp\left[-\frac{\Delta\tau}{2}V(r_m)\right]$, then carry out a Fourier sine transform from space points r_m to points k_n. Multiply that result by the kinetic energy operator $\exp\left(-\Delta\tau\frac{\hbar^2k_n^2}{2m}\right)$, then Fourier transform back to the real-space points, r_j, and finally multiply by $\exp\left[-\frac{\Delta\tau}{2}V(r_j)\right]$. Repeat N times.

While our procedure for producing a discrete representation of the full propagators retains the unitarity property, it is nonetheless an approximation with error terms depending on the spatial grid size. We can show that this particular discretization is especially accurate because (perhaps surprisingly) each matrix product (including the FFT) is equivalent to using trapezoidal rule integration. The Euler-Maclaurin error formula for the

trapezoidal rule [12] states that the asymptotic expansion of the error is proportional to the odd derivatives of the function at the end points, plus a remainder term. All of our end point derivatives are zero to machine accuracy. Thus our grid errors rapidly become negligible once a sufficient number of integration points are employed, about 256 points in our case. Table I shows that spatial discretization errors do not undermine our assertions about the accuracy of extrapolating away time-slice errors.

We now detail the extrapolation in the discretization of "time" to achieve high accuracy and a resulting density matrix that is readily calculated to a precision of 10^{-9} using 64 bit arithmetic. The extrapolation formula of Eq. (5) illustrates the elimination of quadratic errors. Generalizing this is completely equivalent to performing the calculation for several different values of the discretization N, then fitting to a low order polynomial in $1/N^2$ to predict the exact result. We adopt a procedure exactly parallel to the Gear or Bulirsch-Stoer [8] methods for doing polynomial error estimates of integrating differential equations. This procedure involves the successive evaluation of the function for $N = 2, 4, 6, 8, 12, 16, 24, 32, \ldots$ points and a polynomial extrapolation is carried out until a prescribed error tolerance is met. We found it somewhat more efficient to break the full time interval into one to five equal segments, depending on the initial r' value, and in each segment apply the Gear prescription.

For illustrative purposes here, we drop the Gear procedure in order to graphically display the error versus time-step accuracy of the calculation. We start with N time slices, and repeat the calculation with $2N$, $4N$, $8N$, and $16N$ slices. Then Eq. (5) can be easily generalized to eliminate $1/N^2$, $1/N^4$, $1/N^6$, and $1/N^8$ errors. Figure 1 is a plot of the resulting errors in $\rho_0(r, r', \beta)$, for $r' = 3$ Å and $\beta = 0.025/\mathrm{K}$, with $N = 16$ initially. The logarithmic plot clearly illustrates that each removal of an additional power of $1/N^2$ increases accuracy by nearly two orders of magnitude. The ultimate accuracy possible is limited by the word length of the computer and is about half of the accuracy of the hardware. The top curve in the figure also illustrates that normally acceptable accuracy is achievable in calculations with discretizations as coarse as $N = 16$ and $N = 32$ with application of Eq. (5) to remove quadratic errors.

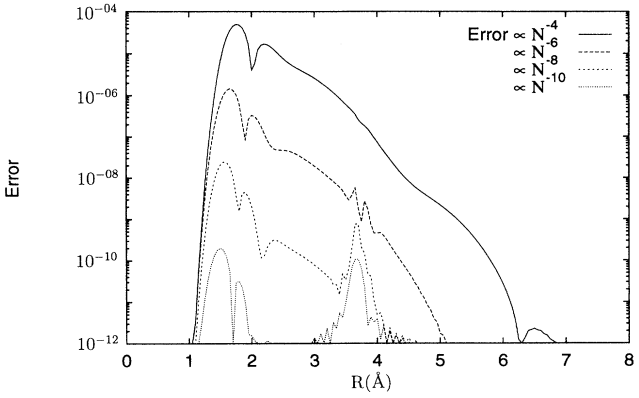

FIG. 1. Error after eliminating successive powers of $1/N^2$.

TABLE I. Comparison of the $l = 0$ radial density matrix $\rho_0(r, r', \beta)$ at $\beta = 0.025/\mathrm{K}$, for various numbers of integration points, M, and step sizes, Δr. The error tolerance for time extrapolations was set at 10^{-9}. All lengths are measured in angstroms. Our value of $\hbar^2/2m$ is 12.119 239 3 Å^2 K.

r	r'	M	Δr	$\rho_0(r, r', \beta)$
2.4	2.0	512	0.05	0.017 939 355
2.4	2.0	512	0.025	0.017 939 355
2.4	2.0	256	0.05	0.017 939 355
2.4	2.0	128	0.10	0.017 939 355
2.4	2.0	64	0.20	0.017 939 142
2.4	2.0	32	0.40	0.017 907 811
2.6	3.2	512	0.05	0.357 527 511
2.6	3.2	512	0.025	0.357 527 511
2.6	3.2	256	0.05	0.357 527 511
2.6	3.2	128	0.10	0.357 527 505
2.6	3.2	64	0.10	0.357 527 500
2.6	3.2	32	0.20	0.357 527 499

In Table I we provide a tabulation of ρ_0 to illustrate the error incurred due to k-space and r-space grid sizes. We see that the grid quickly becomes sufficiently dense that the error in the integration is essentially zero. The remarkable insensitivity of the result to the grid size follows from the Euler-Maclaurin error expansion for the trapezoidal rule.

We have performed the above calculations on other standard two-body He-He potentials, including the Lennard-Jones, hard sphere, and Morse potentials. All assertions in the above analysis remain true as long as the potential is sufficiently repulsive that the density matrix and its derivatives at the origin are small enough to be numerically ignored.

The use of the FFT is not essential to the extrapolation procedure, but because of the special form of the matrix U_{jm} in Eq. (13), the matrix products needed in Eq. (12) can be efficiently carried out. Matrix squaring is an alternative to the FFT, but the computational complexity of these approaches is different. Let N_S be the number of points in the spatial discretization and N_T be the number of terms in the break up formula. Our FFT method requires $N_T N_S^2 \ln N_S$ operations. Matrix squaring requires $N_S^3 \ln N_T$ operations. The N_S^2 in each of the above comes from needing $\rho(r, r', \beta)$ at $N_S \times N_S$ space points. This does not take into account the bandedness of the density matrix. For a given r', the only nonzero values of ρ are for positions r that are within a few thermal wavelengths of r'. (This range is easily estimated assuming free particle propagation.) If we label the number of nonzero values as N_B, then the above estimates change to $N_T N_S N_B \ln N_B$ for FFT, $N_S N_B^2 \ln N_T$ for matrix squaring. In any particular problem, it is likely that FFT will be successful, but a combination of FFT and matrix squaring followed by extrapolation will be optimal.

We have also investigated constructing the density matrix from an eigenfunction expansion, but found this im-

practical except when the eigenfunctions were known analytically. When numerical eigenfunctions are needed, the eigenfunction solver will (it is our experience) introduce unacceptable discretization errors. The complexity for evaluating an eigenfunction expansion requires $N_S^2 N_X$, where N_X is the number of terms needed in the expansion and $N_B N_S N_X$ when bandedness is exploited.

Our primary result is the demonstration that extrapolation to remove time-step errors is highly effective when the error structure of the propagator has only even-order terms. This result may be applied to the matrix squaring method as well. In that case, the cost of doubling the number of time slices is simply the cost of repeating the calculation with one more matrix multiply as compared with the FFT method where doubling the number of time slices actually doubles the cost. If the matrix squaring technique is based on a unitary propagator such as Eq. (3), and the propagator is discretized so that the matrix retains that unitarity, then extrapolation will be very cost effective. We would also point out that the matrix squaring technique can enjoy the same high order accuracy in the spatial grid discretization if the construction of the initial matrix is carried out using a procedure, for instance Eq. (13), where the trapezoidal rule integration scheme and Euler-Maclaurin error estimate applies.

In conclusion, we wish to reiterate that the even-order error structure of the propagator can be utilized by both real- and imaginary-time integration schemes. This allows many fewer time steps or greatly reduces the error for a moderate number of time steps. Our attention was originally drawn to this problem by De Raedt and Michielsen [1], who integrated the Schrödinger equation forward in real time to calculate quantum scattering. That work utilized a result of Suzuki [7] that combined forward and backward time integrations to make the error of a single composite time step fifth order, hence the total error proportional to $1/N^4$. We believe that our method could be used to produce higher-order real-time propagators. However, extrapolations like Eq. (5) produce a propagator that does not in general satisfy the unitarity condition. The violations of the unitary condition is, however, of the same order as the error. This means that our results may require the errors to be extrapolated to high precision to maintain stability for real-time propagations. This is similar to the Tchebychev method [2], which uses a polynomial approximation to the propagator $\exp(-H\Delta\tau)$. This method is also not unitary but the errors and the violation of unitarity are of the same order in $\Delta\tau$ as the order of the highest polynomial.

As our final observation, we note that the extrapolation procedure need not be applied directly to the calculation of an accurate propagator. Some techniques, such as Monte Carlo, do not employ a tabulated representation of the propagator. In such instances, the extrapolation scheme can be used much like standard finite-size scaling methods.

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