Extensions to the distributed approximating functional: The harmonic propagator

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The formalism of the distributed approximating functional for integrating the time-dependent Schrödinger equation is extended to allow an analytic treatment of all Hamiltonians of quadratic order. The resulting propagator matrix is highly banded in both position and momentum representations. For certain choices of the parameters, the action of this matrix can be cast in the form of a Toeplitz operator. We demonstrate that banded Toeplitz matrix-vector multiplication can be performed in significantly fewer numerical operations than is required by conventional split-operator fast Fourier transform algorithms. Numerical examples verify the efficiency of this approach in both storage and speed. Extensions of the method to Monte Carlo path integration, and the implementation on massively parallel computer architectures, are discussed.

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I. INTRODUCTION

The analysis and understanding of dynamical phenomena remains one of the fundamental goals of physics; thus, in recent years, considerable effort has been devoted to solving realistic problems in quantum dynamics. By and large the methods employed can be classified as either time independent or time dependent (although hybrid methods do exist). Time-dependent approaches necessitate finding representations of the quantum propagator which are computationally efficient (in terms of speed) and minimize storage requirements. In recent years effort along these lines has been quite successful as, for example, methods such as the discrete variable representation (DVR) [1], which is also used for the time-independent Schrödinger equation, and fast Fourier transform (FFT) techniques [2] have been successfully applied to systems of a few degrees of freedom, e.g., reactive atom-diatom scattering. Nonetheless, because of the practical limitations (in terms of speed and/or storage requirements) encountered in methods such as DVR, etc., there has been significant interest in exploring new directions, for example, starting from a semiclassical viewpoint [3] or the properties of the Feynman [4] or coherent state [5] path integrals, leading to new formalisms for describing quantum dynamical processes. Our own work follows this second course as, in a recent series of papers, we have developed an entirely new theoretical formalism, that of the distributed approximating functional (DAF) [6–17] which leads to a powerful and efficient algorithm for numerically integrating partial differential equations.

Much of the effort expended in the search for new representations of quantum dynamics has focused on "solving" the time-dependent Schrödinger equation which, unlike the typical time-independent approaches, yields dynamical information without requiring the solution of a large algebraic system of equations. For scattering calculations in the time domain the numerical efficiency of the DAF method can be measured in terms of the number of floating point operations, which scales as N, the number of grid points for dimension $d=1$. This high efficiency is achieved by exploiting the fact that the DAF free propagator $\Delta_S(x,x';t)$ can be cast in a particularly convenient form. That is, $\Delta_S(x,x';t)$ depends on $x-x'$ only; operators of such a functional form are called Toeplitz operators (in discrete terms, a matrix which is constant along the diagonals, i.e., $M_{ij}=M_{i=j}$). For such an operator, matrix-on-vector multiplication can be cast as a convolution and can be performed via the fast Fourier transform in an extremely efficient manner, a method we term the "DAF convolution." As previous work has shown, this fortunate state of affairs holds for the DAF free propagator in any frame moving with constant velocity v. (For $v\neq 0$ we call this the traveling DAF, or TDAF, while the special case $v=0$ we call the stationary DAF or SDAF; hence the subscript on the propagator.) Clearly, for the prototypical scattering calculation, which naturally divides into regions of incoming wave, scattering region, and outgoing wave, constant velocity frames form a logical starting point for describing the dynamics, making the DAF-convolution algorithm the "method of choice" in terms of speed and efficient storage. Moreover, the DAF should prove even more advantageous, relative to FFT techniques, on massively parallel comput-

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er architectures because of the far lower interprocessor communication required by the banded DAF propagator. Studies in this regard are in progress [18] and should be reported shortly.

However, for some classes of physical problems such constant velocity frames bear little relation to the underlying physics of the problem, for example, the dynamics of bound states or "near top of barrier" tunneling. For these cases the dynamics in no way approximates a free wave packet—although it is very similar to the evolution of a packet subject to either a harmonic oscillator or parabolic barrier potential—and as a result the bandwidth of the DAF free propagator must be larger and the time step must be shorter, both reducing the speed of the DAF algorithm and increasing the storage requirements. Alternatively, one can build the localized dynamics (to linear order) into the DAF propagator [15], thus explicitly taking account of the local classical force. This reduces the bandwidth of the DAF propagator and allows for a longer time step, but because the operator is no longer Toeplitz [$\Delta(x,x';t) \neq \Delta(x-x';t)$], each row of the propagator matrix must be calculated and stored separately, and the DAF-convolution algorithm no longer applies. Our studies have shown that using straight banded matrix-on-vector multiplication for wave propagation, the DAF approach is competitive with, but not consistently superior to, split-operator FFT techniques; [16,17] thus, a non-Toeplitz propagator matrix represents a concession in both storage and speed.

Certainly there is nothing numerically special about scattering problems, and given the extremely general nature of the DAF as a means for integrating partial differential equations, one would expect that suitable modifications would allow the DAF formalism to handle other physical problems with the efficiency and speed already demonstrated for scattering. Recent developments in the DAF approach have convinced us this is indeed the case, as the DAF propagator can take into account the underlying dynamics on a gross scale. Thus the efficiencies of the DAF propagator can be extended beyond what has been considered to this point and this paper is the first in a short series detailing some of these extensions.

We begin by considering the prototypical physical cases mentioned above, i.e., bound states and "shallow" tunneling. As noted, the wave packet dynamics for such systems is similar (for sufficiently short time scales) to that of a harmonic oscillator or parabolic barrier potential, respectively. Thus a useful approximation in these cases is to model the system with a suitable quadratic potential, and to treat the dynamics of that potential exactly. This paper shows that the DAF propagator can be generalized to include *analytically* the dynamics of such quadratic potentials, leaving only the nonquadratic terms to be handled numerically. Treating a more representative version of the dynamics exactly allows for longer time steps in the numerical integration and can also lower the bandwidth of the DAF propagator. Moreover, this extension can be accomplished in a way which still yields a Toeplitz DAF propagator matrix—more exactly, the propagation of a wave packet can be reduced to a set of Toeplitz matrix-on-vector multiplications—with all of the associated numerical efficiencies in both speed and storage. Thus, this generalized DAF formalism provides a scheme for integrating the time-dependent Schrödinger equation in a fashion that (i) includes the dynamics exactly up to a global quadratic approximation; (ii) is faster than split operator FFT methods; and (iii) has lower storage and communication requirements.

In addition, although only the simple case of a global quadratic potential is considered here, the nature of our derivation demonstrates it will be possible to find DAF propagators analytically for a much wider class of situations, e.g., the semiclassical evolution of the wave packet. Indeed, it becomes clear that the DAF representation of virtually any transformation mapping one Gaussian wave packet into another—a set of transformations called the metaplectic group [19]—can be calculated analytically. Other useful transformations from this group are planned to be developed in the succeeding papers of this series [20,21].

The development proceeds as follows: Sec. II rederives the DAF formalism in a manner which makes the extensions to quadratic Hamiltonians straightforward. This calculation is carried out explicitly for the useful case of a Hamiltonian of the form $\hat{H}=p^2/2m+m\omega^2x^2/2$ (where ω^2 can be positive, negative, or zero), thus deriving the "harmonic DAF" propagator (HDAF). The results can be cast in two separate, but related, forms, the "progression" HDAF and the "regression" HDAF. (The reason for these names will be clear from the derivation.) Section III examines the specific case of a Toeplitz DAF propagator, develops the DAF-convolution algorithm, and demonstrates it will *in general* be numerically more efficient than conventional split-operator FFT techniques. Although neither the progression nor the regression form of the harmonic DAF propagator is Toeplitz in and of itself, we show that use of the progression and regression propagators in a specific combination does yield an algorithm for wave packet propagation in which all operations are Toeplitz matrix-on-vector multiplies. Thus the harmonic DAF retains the full numerical efficiencies of the DAF convolution approach, even while building in the global quadratic dynamics analytically. We illustrate these points in Sec. IV with the well known example of a wave packet propagating in a Morse potential. Section V concludes with a discussion of results and an overview of work in progress.

II. THE HARMONIC DAF

The DAF arose from a "first principles" effort to create a set of local—in the sense of being confined to a narrow range of coordinate space—"fitting functions" [6] which could be evolved exactly in time under the free particle propagator. Such a set of functions provides a useful means of approximating solutions to partial differential equations, and in particular, the L^2 integrable wave packets commonly encountered in quantum mechanics. As our previous work has shown, the DAF satisfies these fundamental goals as the wave packet can be expressed using the fitting functions and the time evo-

lution can be found in a straightforward manner for any system with a coordinate based potential. Because the fitting functions are localized, the DAF representation of the propagator is banded, allowing the time evolution to be calculated in a numerically efficient fashion.

The DAF representation arises from the replacement of the Dirac delta function $\delta(x)$ by an approximation particularly suited to the purpose at hand, i.e., the integration of an initial value problem. Thus we replace

$$f(x)=\int \delta(x-x')f(x')dx' \tag{1}$$

with the relation

$$f(x)\approx \int \delta(x,x';\mu)f(x')dx' , \tag{2}$$

where $\delta(x,x';\mu)$ is a conveniently chosen (from countless possibilities) approximate representation of the Dirac δ function, and μ denotes the set of parameters used in this approximation. For the propagation of quantum wave functions and/or the calculation of quantum mechanical path integrals a similar approximation can be made to the free particle propagator

$$\begin{aligned}\langle x|e^{-i\hat{p}^2t/2m\hbar}|\Psi\rangle &= e^{-i\hat{p}^2t/2m\hbar}\int \delta(x-x')\Psi(x')dx' \\ &\approx \int [e^{-i\hat{p}^2t/2m\hbar}\delta(x,x';\mu)] \\ &\quad \times\Psi(x')dx' .\end{aligned} \tag{3}$$

For calculating the approximate representation of the free particle propagator a particularly convenient approximation to the δ function is in terms of Hermite polynomials, which represent $\delta(x,x';\mu)$ by

$$\delta(x,x';M,\sigma)=\sum_{n=0}^{M/2} b_n H_{2n}(w)e^{-w^2} , \tag{4}$$

where $w=(x-x')/\sqrt{2\sigma^2}$, σ is a parameter with dimensions of length that determines the "width" of the DAF in coordinate space [6,10], H_j is the jth Hermite polynomial, and

$$b_n=\left[-\frac{1}{4}\right]^n\frac{1}{n!\sqrt{2\pi\sigma^2}} . \tag{5}$$

It should be noted that the DAF can have several other parameters, such as a mean momentum or, as shown below, a characteristic frequency; but, for notational brevity, we will display only μ unless the full parameter set is explicitly needed. In the same spirit of simplicity we utilize a one dimensional notation throughout; generalizations to a higher number of Cartesian coordinates are straightforward (e.g., the DAF propagator matrix is just a direct product) and the generalization to non-Cartesian coordinates has been treated elsewhere [7]. Finally, the product $h_n(w)=H_n(w)e^{-w^2}$ will be termed the nth Hermite function.

The great advantage of the DAF approximation to $\delta(x)$ can be seen by writing the nth Hermite function in terms of a generating function, viz.,

$$H_{2n}(w)e^{-w^2}=\frac{d^{2n}}{dw^{2n}}e^{-w^2} , \tag{6}$$

making it clear that the free particle Hamiltonian, and hence propagator, commutes with the generation of the Hermite function. Moreover, the action of the free particle propagator on e^{-w^2} can be found analytically since this function is itself just the wave function for a Gaussian wave packet.

The Hermite polynomials have several other generating function expressions, and one which proves useful for calculating more general DAF propagators is

$$H_n(w)=\left.\frac{d^n}{d\lambda^n}e^{-\lambda^2+2\lambda w}\right|_{\lambda=0} \tag{7}$$

so that the nth Hermite function is then given by

$$h_n(w)=\left.\frac{d^n}{d\lambda^n}e^{-\lambda^2+2\lambda w-w^2}\right|_{\lambda=0} . \tag{8}$$

The propagator $e^{-i\hat{H}t/\hbar}$ will always be λ independent, and thus commutes with both the differentiation and the limit, i.e.,

$$\begin{aligned}e^{-i\hat{H}t/\hbar}h_n(w) &= e^{-i\hat{H}t/\hbar}\left[\left.\frac{d^n}{d\lambda^n}e^{-\lambda^2+2\lambda w-w^2}\right|_{\lambda=0}\right] \\ &= \left.\frac{d^n}{d\lambda^n}\left[e^{-i\hat{H}t/\hbar}e^{-\lambda^2+2\lambda w-w^2}\right]\right|_{\lambda=0} .\end{aligned} \tag{9}$$

The quantity $e^{-\lambda^2+2\lambda w-w^2}$ can be regarded as a Gaussian wave packet (coherent state wave function) for which propagation under any Hamiltonian with a quadratic potential can be found analytically. In addition, any such propagator transforms a Gaussian wave packet into another Gaussian packet (generally of different width and centered at a different point in phase space) so after the propagator has acted the λ differentiation will still take place on a Gaussian form. Thus the resulting time-propagated DAF can again be written entirely in terms of Hermite polynomials and Gaussian exponentials. Indeed, the derivation leading up to Eq. (9) shows that the functional representation of the DAF in terms of Gaussian exponents and Hermite polynomials is invarient under almost any transformation taking one Gaussian wave function into another one, such as, for example, translations, dilations, or "boosts" of the form $\Psi(x)\rightarrow e^{-ipx/\hbar}\Psi(x)$. The set of all transformations taking one Gaussian wave packet into another is called the metaplectic group [19] and the DAF representation of the group elements can be found straightforwardly provided, of course, the transformation does not depend on λ.

To illustrate the advantages of this generalization to the DAF formalism we explicitly calculate the HDAF propagator $\Delta_H(x,x';t)$ for a DAF boosted to mean momentum $p_0=\hbar k$ (where k is taken to be a constant) in a quadratic potential, i.e., we examine

$$\begin{aligned}\langle x|e^{-i\hat{H}t/\hbar}|\Psi\rangle &= e^{-i\hat{H}t/\hbar}\int e^{ik(x-x')}\delta(x-x') \\ &\quad \times\Psi(x')dx' \\ &= \int [e^{-i\hat{H}t/\hbar}e^{ik(x-x')}\delta(x-x')] \\ &\quad \times\Psi(x')dx' ,\end{aligned} \tag{10}$$

where

$$\hat{H}=\frac{\hat{p}^2}{2m}+\frac{m\omega^2}{2}\hat{x}^2 \tag{11}$$

and ω^2 can be positive, negative, or zero. However, because the evolution operator acts on a function containing $\delta(x-x')$ we are now permitted to make the Hamiltonian instead an operator acting on x', i.e.,

$$e^{-i\hat{H}t/\hbar}e^{ik(x-x')}\delta(x-x')=e^{-i\hat{H}'t/\hbar}e^{ik(x-x')}\delta(x-x') , \tag{12}$$

where

$$\hat{H}'=\frac{\hat{p}'^2}{2m}+\frac{m\omega^2}{2}\hat{x}'^2 \tag{13}$$

and $p'\sim\partial/\partial x'$. Although this interchange makes no formal difference in Eq. (10) it does lead to a different form for the DAF propagator once we approximate the δ function. Thus there are two different (but equally correct) forms for the HDAF propagator, and for reasons which will be apparent below we call the form using $\hat{H}$ the *progression* HDAF and the form using $\hat{H}'$ the *regression* HDAF and denote them by $\Delta_H^P(x,x';t)$ and $\Delta_H^R(x,x';t)$, respectively.

Taking just the progression form for the moment, we have

$$e^{-i\hat{H}t/\hbar}e^{ik(x-x')}\delta(x-x')\approx\Delta_H^P(x,x';t)=e^{-i\hat{H}t/\hbar}e^{ik(x-x')}\delta(x-x';M)$$
$$=\frac{d^n}{d\lambda^n}\left[e^{-i\hat{H}t/\hbar}e^{ik(x-x')}e^{-\lambda^2+2\lambda w-w^2}\right]_{\lambda=0} . \tag{14}$$

The calculation can be made a little more straightforward by remembering a few basic facts about coherent state wavefunctions; that is, consider the coherent state $|z\rangle$ defined by the property that

$$\hat{a}|z\rangle=|z\rangle z ,$$

where

$$\hat{a}=\frac{1}{\sqrt{2}}\left[\frac{\hat{q}}{\sigma}+\frac{i\sigma}{\hbar}\hat{p}\right] \tag{15}$$

and

$$z=\frac{1}{\sqrt{2}}\left[\frac{q}{\sigma}+\frac{i\sigma}{\hbar}p\right] . \tag{16}$$

For such a state the coordinate space wave function is given (up to an overall phase) by

$$\langle x|z\rangle=\left[\frac{1}{\pi\sigma^2}\right]^{1/4}\exp\left[-\frac{(x-q)^2}{2\sigma^2}+\frac{i}{\hbar}p(x-q)\right] . \tag{17}$$

Once again, the parameter σ has dimensions of length, and is a measure of the coordinate space uncertainty of the wave packet. As noted above, for a general quadratic Hamiltonian of the form of Eq. (11), the action of the propagator on the coherent state wave function can be calculated analytically and is given by

$$e^{-i\hat{H}t/\hbar}\langle x|z\rangle=\left[\frac{1}{\pi\sigma^2D_1^2}\right]^{1/4}\exp\left\{\frac{-\sigma^2p^2}{2\hbar^2}+\frac{1}{D_1}\left[\frac{\sigma^2}{2\hbar^2}\left[p-\frac{i\hbar}{\sigma^2}q\right]^2\cos(\omega t)+\frac{ix}{\hbar}\left[p-\frac{i\hbar}{\sigma^2}q\right]-\frac{D_2x^2}{2\sigma^2}\right]\right\} , \tag{18}$$

where

$$D_1=\cos(\omega t)+\frac{i\hbar}{\sigma^2m\omega}\sin(\omega t) \tag{19}$$

and

$$D_2=\cos(\omega t)+\frac{i\sigma^2m\omega}{\hbar}\sin(\omega t) . \tag{20}$$

Note that Eq. (18) is valid for ω^2 positive, negative, or zero. Comparing Eqs. (14) and (17) and recognizing that we will, in general, want $w=(x-x')/\sqrt{2}\sigma$, we see that $e^{-w^2+2\lambda w}$ corresponds (up to an overall constant) to the wave function of a coherent state of width σ centered in position at $q=x'$ and in momentum at $p=\hbar k+\sqrt{2}\hbar\lambda/i\sigma$.

Making use of these identifications in Eq. (18) and applying some algebra the propagation of individual Hermite functions can be written as

$$e^{-i\hat{H}t/\hbar}e^{ik(x-x')}h_n(w)=\rho\frac{d^n}{d\Lambda^n}[e^{-\Lambda^2+2\Lambda\zeta}]_{\Lambda=0}=\frac{1}{\sqrt{D_1}}\left[\frac{\cos(\omega t)}{D_1}\right]^{n/2}e^{i\Delta S/\hbar}e^{-\zeta^2}H_n(\zeta)$$
$$=\frac{1}{\sqrt{D_1}}\left[\frac{\cos(\omega t)}{D_1}\right]^{n/2}e^{i\Delta S/\hbar}h_n(\zeta)\ , \tag{21}$$

where ρ is independent of Λ, and the dummy variable λ has been scaled by

$$\Lambda=\lambda\left[\frac{\cos(\omega t)}{D_1}\right]^{1/2} \tag{22}$$

so that

$$\frac{d^n}{d\lambda^n}=\left[\frac{\cos(\omega t)}{D_1}\right]^{n/2}\frac{d^n}{d\Lambda^n}\ . \tag{23}$$

The final line of Eq. (21) demonstrates the invariance of the DAF propagator alluded to above, i.e., the HDAF propagator can be written entirely (up to overall constants) in terms of Hermite functions. Moreover, this same line shows the strong connection between the quantal DAF propagator and the underlying classical dynamics, for

$$\Delta S=\frac{p_0[x-x'\cos(\omega t)-(p_0/m\omega)\sin(\omega t)]}{\cos(\omega t)}$$
$$+L(p_0,x)\frac{\tan(\omega t)}{\omega}\ , \tag{24}$$

where $p_0=\hbar k$ and the quantity L is the classical Lagrangian [written in first order form, that is, as a function of (q,p) rather than $(q,\dot{q})$] corresponding to the same harmonic potential,

$$L(p,q)=\frac{p^2}{2m}-\frac{m\omega^2}{2}q^2\ . \tag{25}$$

For sufficiently short time $\cos(\omega t)\approx 1$ and $\tan(\omega t)/\omega\approx t$, so ΔS is the change in the classical action for a particle moving in a quadratic potential from x to x'. Similarly, ζ is given by

$$\zeta=\frac{x-x'\cos(\omega t)-(p_0/m\omega)\sin(\omega t)}{\sqrt{2\sigma^2D_1\cos(\omega t)}}\ . \tag{26}$$

Utilizing all of these factors, the progression HDAF propagator is given by

$$\Delta_H^p(x,x';t)=e^{-i\hat{H}t/\hbar}e^{ik(x-x')}\delta(x,x';\mu)$$
$$=e^{i\Delta S/\hbar}\sum_{n=0}^{M/2}c_nH_{2n}(\zeta)e^{-\zeta^2} \tag{27}$$

with

$$c_n=\frac{1}{\sqrt{2\pi\sigma^2D_1}}\frac{1}{n!}\left[-\frac{1}{4}\right]^n\left[\frac{\cos(\omega t)}{D_1}\right]^n\ . \tag{28}$$

Similarly, returning to Eq. (14) and carrying out all of the subsequent steps for the regression form of the propagator yields the same results provided the exchanges $x\rightarrow -x'$, $x'\rightarrow -x$, and $p\rightarrow -p$ are made. Thus the regression HDAF propagator is given by

$$\Delta_H^R(x,x';t)=e^{-i\hat{H}'t/\hbar}e^{ik(x-x')}\delta(x,x';\mu)$$
$$=e^{i\Delta S'/\hbar}\sum_{n=0}^{M/2}c_nH_{2n}(\zeta')e^{-\zeta'^2}\ , \tag{29}$$

where c_n is the same as in the progression version, and now

$$\zeta'=\frac{x\cos(\omega t)-x'-(p_0/m\omega)\sin(\omega t)}{\sqrt{2\sigma^2D_1\cos(\omega t)}}\ , \tag{30}$$

and

$$\Delta S'=\frac{p_0[x\cos(\omega t)-x'-(p_0/m\omega)sin(\omega t)]}{\cos(\omega t)}$$
$$+L(p_0,x')\frac{\tan(\omega t)}{\omega}\ . \tag{31}$$

The origin of the terms progression and regression should now be clear, for in both cases the argument of the Hermite functions, and hence the maximum amplitude (approximately) of the time-propagated DAF, follows the classical path of a particle propagating in a harmonic oscillator of frequency ω,

$$q(t)=q_0\cos(\omega t)+\frac{p_0}{m\omega}\sin(\omega t) \tag{32}$$

(again this formula is valid for all values of ω) but in the former case the argument travels forward in time (progresses) from the phase space point x',p_0 while in the latter case it travels backwards in time (regresses) from the point x,p_0.

Equations (27) and (29) each provide a "basis" in which each element obeys the underlying global quadratic dynamics exactly. Thus, for $\omega^2>0$, either form of the DAF evolves as a periodic function of time with frequency ω. The regression form of the propagator is generally superior to the progression form for numerical purposes (in the case of matrix-on-vector multiplication), and this is particularly true for the two forms of the HDAF; as a rule the progression form of the propagator is far less banded than the regression form. We defer further comparison of the progression and regression propagators to the succeeding paper (in which the DAF propagator is generalized to include the *local* quadratic part of the potential) where it will prove more relevant.

The most important physical parameter in the evolution of the HDAF is the ratio of the "DAF width" σ to the frequency of the harmonic potential ω (and thus to the "natural width" of the potential). Figure 1 depicts an example of a DAF propagating under the effect of a quadratic Hamiltonian (in regression form) for the ratio σ/ω equal to 1. In all cases where $\omega^2>0$ the regression

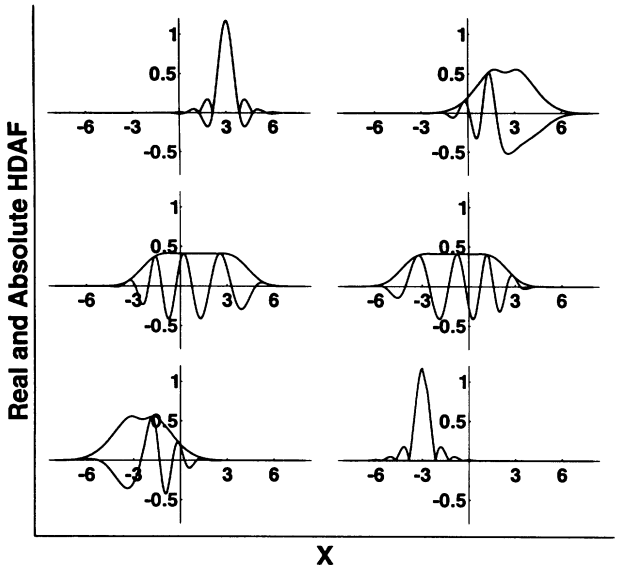

FIG. 1. Time series for the evolution of a DAF under a harmonic oscillator potential (the HDAF propagator) for half a period, in regression form. Here all dimensional parameters $(\sigma,\omega,\hbar,m)$ are set to 1, and the time is in increments of $\pi/5$. The DAF is initially centered at $x'=3$ and the real part (which is identically zero at $t=\pi$) and absolute value are displayed.

HDAF stays localized (indeed, more localized than under the free particle propagator), indicating that the HDAF propagator is banded, and is periodic in time with frequency ω. For $\omega^2<0$, i.e., the parabolic barrier, the propagated DAF is less localized than under the free particle propagator, but this is simply a reflection of the underlying physics, and by treating this spreading analytically we expect the regression HDAF propagator will allow for a longer time step or increased accuracy (or both) to a degree sufficient to make up for the wider bandwidth.

Finally, note that either form of the HDAF propagator represents an extension of much of the DAF formalism presented in earlier works, for taking the appropriate limits on the parameters k and ω in Eq. (27) or (29) yields the propagators for the SDAF and TDAF (Δ_S and Δ_T, respectively). Thus the actions of a general short time propagator (with coordinate based potential) on a wave packet $\Psi(x)$ can be written

$$e^{-i\hat{H}t/\hbar}\Psi(x)\approx e^{-iV_I(\hat{x})t/\hbar}e^{-i\hat{H}_Q t/\hbar}\Psi(x)$$
$$\approx e^{-iV_I(x)t/\hbar}\int \Delta_H(x,x';t)\Psi(x')dx' , \quad (33)$$

where $\hat{H}_Q$ is the Hamiltonian up to quadratic terms, $V_I(\hat{x})$ is the "remainder,"

$$V_I(\hat{x})=\hat{H}-\hat{H}_Q , \quad (34)$$

and Δ_H is the DAF representation of the exact propagator [thus the second line in Eq. (33) represents an additional level of approximation since the DAF cannot exactly reproduce any L^2 integrable wave packet]. Note that because the DAF provides a coordinate space representation the operator $e^{-iV_I(\hat{x})t/\hbar}$ leads only to a scalar multiplication, as the notation of the last line of Eq. (33) indicates. For somewhat greater accuracy, a symmetric Trotter splitting can be used instead of that in Eq. (33), yielding

$$e^{-i\hat{H}t/\hbar}\Psi(x)\approx e^{-iV_I(\hat{x})t/2\hbar}e^{-i\hat{H}_Q t/\hbar}e^{-iV_I(\hat{x})t/2\hbar}\Psi(x)$$
$$\approx e^{-iV_I(x)t/2\hbar}\int \Delta_H(x,x';t)e^{-iV_I(x')t/2\hbar}\Psi(x')dx' , \quad (35)$$

which has errors of $O(\epsilon^3)$ as opposed to the $O(\epsilon^2)$ ones of Eq. (33). Alternatively, one can make use of other forms of operator splitting, such as the modified Cayley approach [22].

III. THE EFFICIENCIES OF THE DAF-CONVOLUTION APPROACH

In two recent calculations of time-dependent scattering amplitudes [16,17] we have demonstrated that the DAF approach can be both faster (in terms of CPU time) and have lower storage requirements than conventional split-operator FFT methods. These results made use of a specific algorithm, termed the DAF convolution, the efficiencies of which hinged upon the fact that the discrete SDAF or TDAF propagator matrix is a Toeplitz operator, and thus the matrix-vector product involved in propagating the wave packet can be computed via a convolution. This section develops the theoretical underpinnings of the DAF-convolution algorithm and shows it will *in general* be numerically more efficient than split-operator FFT methods. Although the HDAF propagator cannot be cast entirely in Toeplitz form, it is possible to evolve wave functions under the HDAF via *a set* of Toeplitz operators, thus maintaining the significant advantages in speed and storage of the Toeplitz operator structure.

For the purpose at hand it is sufficient to consider the simplest form of DAF propagator which is Toeplitz; thus, consider the action of the SDAF propagator $\Delta_S(x,x';\tau)$ on a one dimensional wave packet propagating freely for time τ

$$\psi(x;t+\tau)=\int \Delta_S(x,x';\tau)\psi(x';t)dx' ,$$

where the SDAF propagator is given by taking $k=\omega=0$ in Eq. (27) or (29)

$$\Delta_S(x,x';t)=\sum_{n=0}^{M/2} b_n\left[\frac{\sigma}{\sigma(t)}\right]^{n+1} H_{2n}\left[\frac{x-x'}{\sqrt{2\sigma^2(t)}}\right]$$
$$\times\exp\left[-\frac{(x-x')^2}{2\sigma^2(t)}\right] , \quad (36)$$

which is explicitly Toeplitz [i.e., $\Delta_S(x,x';t)=\Delta_S(x-x';t)$]. The discrete representation of Δ_S will be banded, and we take the bandwidth to be $2w+1$. Assume, for ease of exposition, that there are $n=km$ grid points, with $m \geq w$. Write Δ_S as

$$\Delta_S = \begin{bmatrix} \mathbf{A} & \mathbf{B} & 0 & \cdots & 0 \\ \mathbf{B}^T & \mathbf{A} & \mathbf{B} & & \vdots \\ 0 & \mathbf{B}^T & \mathbf{A} & & 0 \\ 0 & 0 & & \ddots & \mathbf{B} \\ 0 & 0 & \cdots & \mathbf{B}^T & \mathbf{A} \end{bmatrix} , \tag{37}$$

where now $\mathbf{A}$ is a dense (i.e., not banded) $m \times m$ symmetric Toeplitz matrix, $\mathbf{B}$ is Toeplitz, lower triangular and $m \times m$, and $\mathbf{B}^T$ is the transpose of $\mathbf{B}$.

If we partition ψ conformably we have

$$\Delta_S \psi = \begin{bmatrix} \mathbf{A}\psi_1 + \mathbf{B}\psi_2 \\ \mathbf{B}^T\psi_1 + \mathbf{A}\psi_2 + \mathbf{B}\psi_3 \\ \vdots \\ \mathbf{B}^T\psi_{k-1} + \mathbf{A}\psi_k \end{bmatrix} . \tag{38}$$

The banded Toeplitz matrix-vector product is thus reduced to a set of $3k-2$ smaller Toeplitz matrix-vector products. Each of the products $\mathbf{A}\psi_j$, $\mathbf{B}\psi_j$, or $\mathbf{B}^T\psi_j$ times ψ can be computed in $O(m \log_2 m)$ flops, accomplished via a fast fourier transform, which need be applied to $\mathbf{A}$ and $\mathbf{B}$ only *once* for the entire propagation sequence.

To see how this works, notice that the FFT can be viewed simply as a fast method for computing the convolution of two vectors. The convolution of the (discrete, n periodic) vectors is the vector quantity $z=x \circ y$, defined by $z_k=\sum_{j=0}^{n-1} x_j y_{k-j}$ (this product can represent, e.g., the product of two polynomials). The convolution of x and y can also be written as the matrix-vector product $z=\mathbf{X}y$, where $\mathbf{X}$ is the circulant, or convolution, matrix (written here as a set of column vectors)

$$\mathbf{X}=[x,\mathbf{S}x,\mathbf{S}^2x,\ldots,\mathbf{S}^{n-1}x] ,$$

and $\mathbf{S}$ is the downshift matrix (again, as a set of column vectors)

$$\mathbf{S}=[\mathbf{e}_2,\mathbf{e}_3,\ldots,\mathbf{e}_n,\mathbf{e}_1]$$

with $\mathbf{e}_j$ equal to the jth column of the $n \times n$ identity matrix (see, e.g., [23]).

Now the discrete Fourier matrix $\mathbf{F}$ diagonalizes every circulant matrix, and thus $x \circ y = \mathbf{X}y = \mathbf{F}\mathbf{D}\mathbf{F}^{-1}y$ can be computed (by an FFT) in $O(n \log_2 n)$ operations. The application of this theory to the action of a finite Toeplitz matrix is achieved by noticing that a Toeplitz matrix of order m can always be embedded in a circulant matrix of order M, with $M \geq 2m-1$. Thus, for example, we can choose P, Q, and R such that the $M \times M$ matrix

$$\tilde{\mathbf{A}} = \begin{bmatrix} \mathbf{A} & \mathbf{P} \\ \mathbf{Q} & \mathbf{R} \end{bmatrix} \tag{39}$$

is circulant. Then by taking the product

$$\tilde{\mathbf{A}} \begin{bmatrix} y \\ 0 \end{bmatrix} = \begin{bmatrix} \mathbf{A}y \\ \mathbf{Q}y \end{bmatrix} , \tag{40}$$

the desired quantity ($\mathbf{A}y$) can be easily extracted. Simple algebra shows that using this method, the action of a Toeplitz matrix of order n with bandwidth $2w+1$ can be calculated in $O(n \log_2 w)$ flops.

Comparison with Eqs. (26) and (30) shows that neither the progression nor the regression form of the HDAF propagator is Toeplitz; however, each can be written as the product of a scalar operator with an operator that is "almost Toeplitz," i.e., operators with arguments going like $x-x'\cos(\omega t)$ and $x\cos(\omega t)-x'$, respectively. That is, the progression HDAF propagator can be written

$$\Delta_H^P(x,x';t) = \exp\left[-\frac{i}{\hbar} \frac{m\omega x^2}{2} \tan(\omega t) \right] \times F(x-x'\cos(\omega t);p,t) \tag{41}$$

and similarly for the regression version

$$\Delta_H^R(x,x';t) = \exp\left[-\frac{i}{\hbar} \frac{m\omega x'^2}{2} \tan(\omega t) \right] \times F(x'-x\cos(\omega t);p,t) , \tag{42}$$

with the function F given by

$$F(y;p,t) = \exp\left[\frac{i}{\hbar} \left[\frac{py}{\cos(\omega t)} - \frac{p^2 \tan(\omega t)}{2m\omega} \right] \right] \times \sum_{n=0}^{M/2} c_n h_{2n} \left[\frac{y-(p/m\omega)\sin(\omega t)}{2\sigma^2 D_1 \cos(\omega t)} \right] . \tag{43}$$

By using the progression and regression forms in combination the HDAF propagator can be reduced to a set of Toeplitz matrix-on-vector and scalar update operations. To wit: consider the coordinate space representation of the evolution operator

$$\begin{aligned} \langle x|e^{-i\hat{H}t/\hbar}|x'\rangle &= \int \langle x|e^{-i\hat{H}t/2\hbar}|x''\rangle\langle x''|e^{-i\hat{H}t/2\hbar}|x'\rangle dx'' \\ &\approx \int \Delta_H^R(x,x'';t/2) e^{-iV_I(x'')t/2\hbar} e^{-iV_I(x'')t/2\hbar} \Delta_H^P(x'',x';t/2) dx'' \\ &= \int F(x''-x\cos(\omega t))F(x''-x'\cos(\omega t))\Gamma(x'')dx'' , \end{aligned} \tag{44}$$

where x'' represents an intermediate coordinate eigenstate and

$$\Gamma(x'')=\exp\left[-\frac{i}{\hbar}[2tV_I(x'')+m\omega x^2\tan(\omega t)]\right] .$$

From Eqs. (41) and (42), the change of variable

$$x''=y\cos(\omega t) \tag{45}$$

makes both factors of F in the last line of Eq. (44) Toeplitz, and since Γ is a scalar function, the HDAF propagator for two time steps has been reduced entirely to Toeplitz operations.

After the above change of variables the discrete form of Eq. (44) can be interpreted as mapping the wave packet between two grids, the first taking the packet from the regular coordinate state grid on which the DAFs are placed to a second intermediate grid scaled by $\cos(\omega t)$. The second time step then maps the packet back onto the "physical" grid. Clearly, the change of variables in Eq. (45) will affect the numerical stability of the HDAF propagation and thus its efficiency compared with other methods, particularly as $\omega t\rightarrow(j+1/2)\pi$ where the transformation becomes noninvertible. Since this series of papers are basically "developmental" in nature, we limit ourselves for the moment to demonstrating that the Toeplitz HDAF propagator accurately integrates the time-dependent Schrödinger equation, deferring a detailed numerical investigation of the HDAF and its efficiencies to the treatment of a suitably *apropos* application.

IV. A SIMPLE NUMERICAL EXAMPLE

In the succeeding papers of the series it will prove convenient to have a common model problem for which some reasonable portion of the dynamics can be expressed analytically; the Morse oscillator is one of the few anharmonic systems for which this is possible, and thus we digress briefly and examine it in detail.

The Morse potential takes the form

$$V(x)=a[(1-e^{-bx})^2-1] , \tag{46}$$

and we note several features of this potential: (i) the minimum lies at $-a$; (ii) the zeros of the potential occur at $x=\ln 2^{-b}$ and as $x\rightarrow\infty$; (iii) there is an exponentially growing hard wall for negative x; and (iv) the well "opens up" much faster than the harmonic oscillator potential found by taking a Taylor expansion around the minimum, so the period will in general be much longer than the period for that oscillator, and becomes infinite as $E\rightarrow 0$.

As noted above, an analytic solution can be found for a classical particle moving in this potential. Consider the case in which the classical particle starts at the phase point x_0,p_0 and has energy

$$E=\frac{p_0^2}{2}+a[(1-e^{-bx_0})^2-1] ,$$

i.e., the mass m has been set to 1. Define the three auxiliary quantities

$$\alpha=\frac{a}{E}+e^{bx_0} ,$$

$$\beta=\frac{p_0}{\sqrt{2E}}e^{bx_0} ,$$

$$\gamma=-\frac{a}{E} ,$$

as well as the quantities

$$\begin{aligned} A(t)&=\alpha\cosh(\nu t)+\beta\sinh(\nu t)+\gamma ,\\ B(t)&=\alpha\sinh(\nu t)+\beta\cosh(\nu t)=\frac{1}{\nu}\frac{dA(t)}{dt} . \end{aligned} \tag{47}$$

Then one can easily show (for example, by substituting into Newton's second law) that

$$x(t)=\frac{1}{b}\ln A(t) , \tag{48}$$

$$p(t)=\sqrt{2E}\,\frac{B(t)}{A(t)} , \tag{49}$$

where

$$\nu=b\sqrt{2E} \tag{50}$$

gives the classical frequency of the oscillation. Note that if $E<0$ then $\nu\rightarrow i\omega$ where $\omega=b\sqrt{-2E}$. In that case

$$\beta\rightarrow\frac{ip_0}{\omega}e^{bx_0}$$

so that $A(t)$, and hence $x(t)$ stays real. Note also that

$$\lim_{t\rightarrow 0}x(t)=\frac{1}{b}\ln(\alpha+\gamma)=\frac{1}{b}\ln e^{bx_0}=x_0$$

as expected. Thus the classical frequency of such a Morse oscillator is $\omega=b\sqrt{-2E}$ and, for example, this frequency can be built in to the HDAF dynamics analytically.

For a specific numerical example we take $a=10.25$ and $b=0.2209$ in the potential of Eq. (46) and propagate a wave packet of unit width ($\sigma=0$) initially located on the soft wall of the potential, corresponding to a coherent state centered at the phase space point $p=0$, $q=5.0$. With this choice of parameters the initial "energy" of the packet—determined by using the expectation values of the position and momentum in the classical Hamiltonian—is $E_0=-5.67$ and the corresponding classical frequency, obtained from Eq. (50), is $\omega_c=0.744$.

Figure 2 shows the phase space trajectory of the center of the wave packet—which is the trajectory of the corresponding classical system—as well as the initial configuration. Figure 3 shows the "survival probability" $P(t)=|C(t)|^2$ where $C(t)$ is the quantum autocorrelation function

$$C(t)=\langle\Psi_0|e^{-i\hat{H}t/\hbar}|\Psi_0\rangle \tag{51}$$

calculated using the HDAF propagator of Eq. (44) with $\omega=\omega_c$. Although, as noted above, we defer an in-depth examination of the numerical properties of the HDAF until treating a "real world" problem, is it noteworthy that the accuracy of the results is not extremely sensitive

to the value taken for ω. That is, while "building in" the underlying bound state dynamics aids in the wave packet propagation, accurate results can still be obtained—usually at the cost of a shorter time step—by utilizing a frequency other than the classical one.

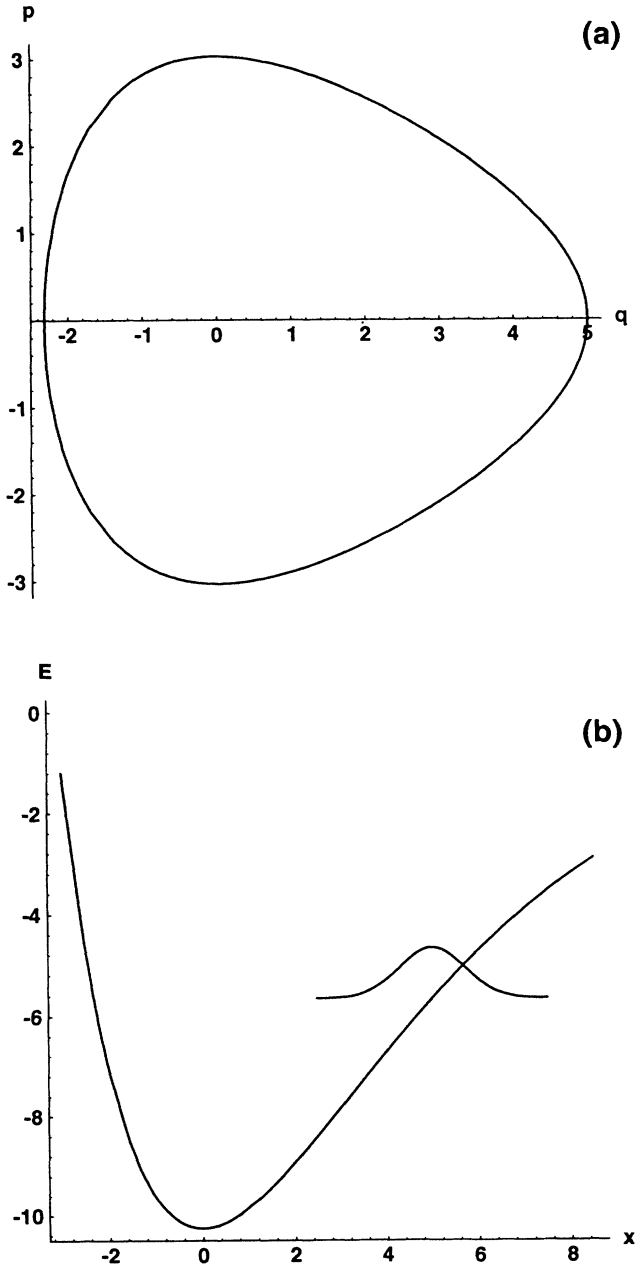

FIG. 2. Some of the salient features of the simple numerical example presented in this section: (a) The classical phase space trajectory (the trajectory followed, to first order in $\hbar$, by the center of the wave packet) corresponding to the Morse potential with the given parameters. (b) The initial configuration of the system, with the Gaussian wave packet centered at $q_0=5$ and having no initial momentum. The packet is shifted vertically to reflect the classical energy of $E=-5.67$.

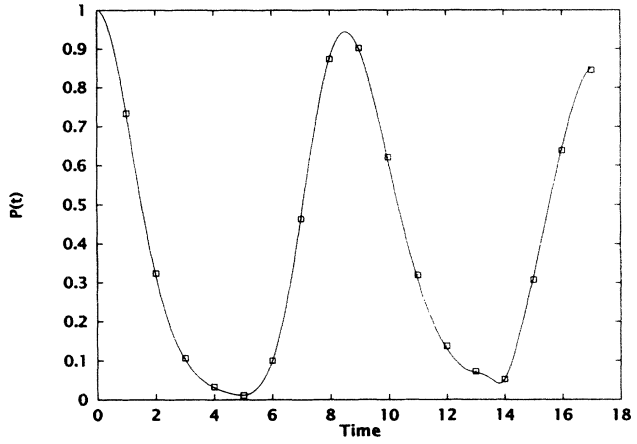

FIG. 3. The survival probability $P(t)$ for the Gaussian packet evolving in the Morse potential. The exact results (solid line) were obtained via a converged SDAF calculations (itself checked with a split operator FFT algorithm) while the discrete points were calculated using the progression and regression HDAF propagators in the fashion described in Sec. IV, i.e., wave packet propagation is reduced to a set of Toeplitz matrix-on-vector operations which, via the DAF convolution, means the total work scales as $O(N)$. The frequency of the harmonic potential in the HDAF propagators was set to the frequency of a particle of the given energy in the corresponding classical Morse oscillator.

V. CONCLUSIONS

This paper has extended the power of the DAF formalism in several ways.

(1) We have explicitly proven a fact recently demonstrated numerically [16,17], i.e., that the DAF-convolution formalism is computationally more efficient than split-operator FFT methods, requiring fewer floating point operations for a given problem. The DAF formalism also has lower storage requirements and, when implemented on massively parallel computers, lower communication costs. All of the above comparisons assume the same number of grid points per degree of freedom for the DAF and the split-operator FFT algorithm; our experience shows the DAF generally requires *fewer* grid points than the FFT, providing the DAF with even greater relative efficiency. In fact, recent results indicate it may be possible to utilize the DAF with far fewer grid points than required by FFT based algorithms. This would prove crucial in the simulation of quantum systems with many degrees of freedom, not just because of the increased speed of the algorithm, but also because of the tremendous reduction in storage requirements for the multidimensional wave function.

(2) We have demonstrated the DAF propagator formalism can include *analytically* the effects of the Hamiltonian through the quadratic terms in the potential.

(3) The major efficiencies in terms of speed of the DAF-convolution algorithm are maintained, as are the lower communication costs and storage requirements, *provided* the wave packet is propagated on a uniform grid and the quadratic approximation to the Hamiltonian is

taken to be global.

(4) From the derivation leading to the HDAF propagator, in particular Eq. (9), analytic DAF propagators can be found for an extremely wide range of transformations, including those of the metaplectic group.

As the derivation of the HDAF makes clear, DAFs in many ways play a role akin to a set of basis functions, for they provide a means of representing functions and operators. Indeed, DAFs constitute a localized, nonorthogonal, basis. However, it must be stressed that we *never* use the DAF as a basis set in the conventional sense; i.e., we never write

$$|\psi\rangle = \sum_n a_n |\phi_n\rangle ,$$

where

$$a_n = \sum_m S^{-1}_{nm} \langle \phi_m | \psi \rangle$$

and $S_{mn} = \langle \phi_m | \phi_n \rangle$ is the "overlap matrix" of the basis vectors. Instead, we utilize the DAF as a set of linear functionals mapping between two different, approximate, representations of the wave packet. For these reasons we refer to the entire DAF method for propagating wave packets as a "non-basis-set approach" to quantum mechanics.

Thus, through DAFs one achieves perhaps the nicest aspects of a coherent state representation, i.e., locality and analytic evolution under a class of Hamiltonians, without having to deal with the technical and numerical difficulties present with coherent states, such as phase space representations of operators or diagonalization of the (pseudosingular) overlap matrix corresponding to an overcomplete nonorthogonal basis. (By pseudosingular we mean a matrix which for any finite size cannot formally have a zero eigenvalue, but some subset of the eigenvalues will be extremely—in the case of coherent states, exponentially—small and the matrix is singular in the $N \to \infty$ limit.) As the derivation of the HDAF propagator made clear, there are strong similarities between DAFs and coherent states, and in many ways we regard the DAFs as a basis of "dressed" coherent states (usually of a single momentum) but we stress again, the DAF is not a type of coherent state (nor is it a wavelet). In the near future we will present a direct comparison between DAFs and basis set expansions demonstrating, in fact, that basis set expansions are a subset of DAFs [24].

These results leave us convinced that the DAF approach holds significant promise for resolving the quantum dynamics problem, for the DAF provides all the advantages of a localized basis set, but are far more "flexible" than the conventional methods of quantum dynamics, allowing (as the current paper has shown) exact treatment of a wide range of transformations, or, for example, nonuniformity of the coordinate space grid or position dependence of the relevant parameters.

Nonetheless, the efficiencies of the DAF-convolution algorithm are obtained at the cost of using a uniform grid in each of the coordinates. Throughout the development of the DAF formalism we have striven to take the "long view" regarding the quantum dynamics problem; that is, we seek a general method for simulating the dynamics of arbitrary—within the limitation that thus far we have considered only the local potentials commonly encountered in chemical physics—quantum systems of several degrees of freedom, where "several" means any dimension such that statistical averaging does not yield a sufficiently good approximation to simplify the problem. Thus, although the extension of the DAF-convolution approach to include the exact global quadratic dynamics greatly widens the class of problems which can be treated in an efficient manner—indeed, we believe this approach brings an entirely new class of problems within computational reach—it does not, in and of itself, resolve our long range goals. There are two principal reasons for this statement: First, for many interesting physical problems, e.g., systems with multiple minima, a global quadratic approximation is of limited use as the convexity of the potential does not even approach being a constant in space. Second, even though the DAF approach has extremely low storage requirements compared to other methods, as the number of degrees of freedom rises, storage *must* become a limiting factor for any method which propagates wave packets directly since wave functions scale geometrically in terms of storage [25]. While not eliminating either of the limitations noted above, we believe the extended DAF formalism presented here points the way to their resolution. Specifically, as with previous work using the stationary or traveling DAF, the global quadratic approximation in the harmonic DAF can instead by made "local," either by using more than one grid [11] or by having each DAF incorporate the *local* potential through quadratic order.

On multiple grids the full DAF propagator will not be Toeplitz, and the DAF-convolution algorithm cannot be applied; however, the propagator matrix will be composed of Toeplitz submatrices corresponding to each grid (with "boundary regions" mapping between them) for which the DAF-convolution does hold. Thus, by using a patchwork of several grids, each with a "semilocal" quadratic approximation to the potential, most of the high-speed and low-storage advantages of the DAF convolution approach can be retained while still treating much of the potential analytically through quadratic order (although the issue of storing the wave function and the propagator matrix for many degrees of freedom remains).

Alternatively, one can imagine propagating each DAF independently using parameters based on some local quadratic approximation to the potential. While this removes any possibility that the DAF propagator matrix will be Toeplitz, it makes way for other savings which may prove more important for large dimensional systems; in particular, it (a) offers an appealing approach to numerical path integration via importance sampling techniques and (b) opens the way for using the DAF as a "dynamical basis set" arrayed along the semiclassical trajectory in phase space, with each DAF evolving exactly under the local quadratic dynamics. Both of these approaches have shown significant promise in the past [3–5] and thus, in a succeeding paper, we develop a collection of local quadratic DAF propagators.

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[25] As the physicist L. S. Brown is fond of pointing out, "There are not enough atoms in the universe to make the paper to write down the wave equation for a uranium atom."