Unitary integration with operator splitting for weakly dissipative systems

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Abstract
Unitary integration is a numerical method that preserves the structure of the quantum Liouville equation by evolving the density via unitary transformations. Unitary integrators preserve the kinematic invariants $c_j = \mathrm{tr}\,\rho^j$ ($j = 1, \ldots, n$) to all orders in the time step. Here we extend unitary integration to weakly dissipative systems. We apply the technique of operator splitting, using a unitary integrator for the Hamiltonian evolution and a conventional integrator for the dissipative piece. In this way, we guarantee that all dissipation and decoherence (variation of the c_j) is due to the new non-Hamiltonian terms and not to any numerical artifacts. We illustrate the method with examples.

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1. Introduction

It is common that the differential equations used to model physical systems possess some 'structural' features that embody important physical properties (e.g., differential-algebraic invariants such as energy) which are reflected in the qualitative behaviour of the system. Often, this structure is purposely built-in to the model on physical grounds. Unless the numerical methods are specially designed, a numerical solution of such a system will not, in general, respect this structure, leaving open the possibility of qualitatively incorrect results. Structural considerations are particularly important when the time domain of interest is much larger than the system's characteristic time scale(s). In such cases it can be difficult, if not impossible, to obtain even qualitatively correct results when the numerical method does not preserve the system's structure. (A particularly interesting example is the simulation of the long-time evolution of the solar system where phase-space conserving (symplectic) techniques proved indispensable [1].) At the very least, the penalty for ignoring the essential structure of the system is a greatly increased computational cost forced by the necessity of a small time step to keep the effects of structural errors from accumulating catastrophically. For a more detailed

discussion of these effects along with various examples see Shadwick *et al* [2] and the references therein.

The goal is not to eliminate all numerical error but rather to identify and minimize those classes of numerical errors that are most damaging to the solution. Backwards error analysis [3–7] provides significant understanding in how a numerical method may lead to a numerical solution with qualitatively incorrect behaviour. The basic approach is to take the view that when a numerical method is applied to a given differential equation, the resulting numerical solution, while an approximation to the exact solution of the original equation, is, in fact, the exact solution of *some* differential equation. Clearly this second differential equation is related to the original equation, and the form of this new equation can often lead to valuable insight into the behaviour of the numerical method [8]. Of particular importance will be the physical consequence of any coupling terms not present in the original system; it is terms of this type which are the cause of structural errors.

These ideas are also relevant to weakly non-ideal systems, such as systems with a small amount of dissipation. Using the technique of operator splitting [9, 10], one separates the differential operator into two pieces: the operator representing the ideal part of the system and whatever non-ideal terms remain. A structure-preserving integrator is used for the former and a generic method for the latter. This technique guarantees that the numerical solution has the proper limit as the dissipation terms vanish. Specifically, consider the system of equations

$$\dot{\psi} = \mathcal{L}_1[\psi] + \mathcal{L}_2[\psi] \tag{1}$$

and let $\mathcal{S}_1(\tau)$ and $\mathcal{S}_2(\tau)$ be the respective numerical evolution operators for a time interval τ. If $\mathcal{S}_1$ and $\mathcal{S}_2$ are accurate to at least second order, then a second-order approximation to the full evolution is

$$\psi(t+\tau) = \mathcal{S}_1(\tfrac{1}{2}\tau) \circ \mathcal{S}_2(\tau) \circ \mathcal{S}_1(\tfrac{1}{2}\tau)\psi(t) + \mathcal{O}(\tau^3) \tag{2}$$

where $\circ$ denotes operator composition.

Consider a weakly dissipative system, which in the absence of dissipation possesses some structure (for example, in the ideal limit the system may be Hamiltonian). In this case, the structural errors committed by standard methods can mimic the effects of dissipation, enhancing (or perhaps masking) what would otherwise be a small effect. In strongly non-ideal systems, the errors introduced in evolving the ideal part of the system are not as significant in that they are not likely to yield behaviours that are not otherwise present. The same is true for errors encountered in the evolution of the dissipative terms. Empirically, one finds that dissipation terms tend to be sufficiently simple that they possess no delicate structure. Consequently, numerical errors will generally not result in qualitatively different behaviour, merely in different effective values for the dissipation parameters.

2. n-level quantum systems with weak dissipation

Our interest here is to explore how these ideas can be applied to an n-level quantum system described by a density matrix, ρ, subject to both reversible and irreversible processes. A general dynamical equation governing the evolution of ρ is the master equation [11]

$$\hbar\dot{\rho} = -\mathrm{i}[H, \rho] + \Lambda[\rho] \tag{3}$$

where the hermitian Hamiltonian H describes the reversible dynamics and the (general) dissipation[3] is represented by the linear operator Λ. Examples include an atomic system

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[3] Although here we are primarily concerned with dissipative processes, Λ can represent arbitrary non-Hamiltonian processes.

interacting with an external radiation field subject to spontaneous emission or a collection of ions in a trapping potential subject to external laser pulses and decoherence. We are most concerned with the case where the irreversible processes are weak compared to the reversible processes, i.e., the case $\|\Lambda\| \ll \|H\|$.

For the moment, let us consider the properties of the quantum Liouville equation [12], the ideal counterpart of (3):

$$\hbar \dot{\rho} = -\mathrm{i} [H, \rho]. \tag{4}$$

This equation has a non-trivial kinematic structure—the Hioe–Eberly invariants

$$c_j = \mathrm{tr}\, \rho^j \qquad j = 1, 2 \ldots, n \tag{5}$$

are non-evolving regardless of the form of the Hamiltonian [13]. These invariants are a direct consequence of the unitary evolution of the density matrix and are the analogues of the Poincaré invariants in classical mechanics, carrying information of equal physical import. A numerical solution of (4) where the c_j are not preserved is thus in danger of being unphysical.

In the method of unitary integration [14], the structure of the quantum Liouville equation is preserved exactly since the numerical time advance map is constructed as a unitary transformation. That is, the density matrix is evolved through a time interval τ according to

$$\varrho_{n+1} = U \varrho_n U^{\dagger} \tag{6}$$

where ϱ_n is the numerical approximation to $\rho(n\tau)$ and U is a unitary matrix which approximates the true evolution operator $\mathfrak{U}(t, t + \tau)$ accurate through order κ in τ:

$$U = \mathfrak{U}(t, t + \tau) + \mathcal{O}(\tau^{\kappa+1}). \tag{7}$$

Thus, although ϱ_n is only a numerical approximation to the true solution of the quantum Liouville equation, the c_j are exactly conserved. Furthermore, since the numerical method advances ϱ_n in time by a unitary transformation, we are guaranteed that the numerical solution will be hermitian.

Returning to (3), our strategy is to use a unitary integrator for the Hamiltonian part of the evolution and a generic algorithm for the dissipative terms. Thus in the limit $\|\Lambda\| \to 0$, the Hioe–Eberly invariants are exactly conserved. In this way we guarantee that all dissipation and decoherence (variation of the c_j) are due to the non-Hamiltonian terms, and not to any numerical artifacts.

3. Constructing unitary integrators

As with every unitary matrix, U can be expressed as the exponential of an anti-hermitian matrix. For our purposes, it is convenient to write

$$U(t, \tau) = \mathrm{e}^{-\mathrm{i}\tau A(t,\tau)} \tag{8}$$

where A is hermitian. The matrix A is computed by expanding (6) in a Taylor series about $\tau = 0$ and matching term-by-term with the Taylor series for $\rho(t + \tau)$ obtained from the quantum Liouville equation. A straightforward, if tedious, calculation [14] reveals the following approximations for A:

to second order

$$A = H(t) + \frac{1}{2!} \tau H'(t); \tag{9}$$

to third order

$$A = H(t) + \frac{1}{2!} \tau H'(t) + \frac{1}{3!} \tau^2 H''(t) + \frac{i}{12} \tau^2 \left[H(t), H'(t) \right]; \tag{10}$$

and to fourth order[4]

$$A = H(t) + \frac{1}{2!} \tau H(t)' + \frac{1}{3!} \tau^2 H(t)'' + \frac{1}{4!} \tau^3 H(t)''' + \frac{i}{12} \tau^2 \left[H(t), H(t)' \right]$$

$$+ \frac{i}{4!} \tau^3 \left[H(t), H(t)'' \right]. \tag{11}$$

Note that to obtain accuracy beyond second order, one must take into account that, in general, $[H(t_1), H(t_2)] \neq 0$.

Fortunately, to use these expressions for U, it is not necessary to exponentiate a (general, possibly quite dense) $n \times n$ matrix. We are free to approximate A in any way consistent with the order of the method. This freedom can be used to simplify the construction of $U(t, \tau)$. In the following, it is convenient to assume that H is traceless. To begin, let λ_k be a basis for the set of all (traceless) $n \times n$ hermitian matrices, with normalization

$$\text{tr } \lambda_i \lambda_j = 2 \delta_{ij} \tag{12}$$

whence

$$A = \sum_{k=1}^{n^2-1} \alpha_k \lambda_k \qquad \text{where} \qquad \alpha_k = \frac{1}{2} \text{tr } A \lambda_k. \tag{13}$$

In addition, we may choose the λ_k in such a way that either λ_k is diagonal or has at most two non-zero elements. In either case, $e^{ia\lambda_k}$ is easily computed. Our goal is to replace the single exponential in (8) with a product of exponentials of the λ_k:

$$e^{-i\tau A} = \prod_{k=1}^{n^2-1} e^{-i\tau \beta_k \lambda_k} + \mathcal{O}(\tau^{\kappa+1}) \tag{14}$$

for some β_k. Since the λ_k do not necessarily commute with one another, the β_k will depend on the α_j in a complicated manner.

While it is possible to determine the β_k in (14) by repeated application of either the Campbell–Baker–Hausdorf [15–17] or the Zassenhaus [18] formula, such an approach quickly becomes quite complicated as n increases and does not seem to be well suited to symbolic computation. A more straightforward approach is, using (13), to expand the left- and right-hand sides of (14) as polynomials in τ and match terms order-by-order [19]. For an integrator of order κ, we can write α_k as

$$\alpha_k = \sum_{j=0}^{\kappa-1} \tau^j \alpha_k^{(j)} \tag{15}$$

and similarly

$$\beta_k = \sum_{j=0}^{\kappa-1} \tau^j \beta_k^{(j)} + \mathcal{O}(\tau^{\kappa}). \tag{16}$$

Using these expressions in (14), we obtain

$$1 - i \tau \sum_{j=0}^{\kappa-1} \sum_{k=1}^{n^2-1} \tau^j \alpha_k^{(j)} \lambda_k - \frac{1}{2} \tau^2 \sum_{j,j'=0}^{\kappa-1} \sum_{k,k'=1}^{n^2-1} \tau^{j+j'} \alpha_k^{(j)} \alpha_{k'}^{(j')} \lambda_k \lambda_{k'} + \ldots$$

$$= \prod_{k=1}^{n^2-1} \left(1 - i \tau \sum_{j=0}^{\kappa-1} \tau^j \beta_k^{(j)} \lambda_k - \frac{1}{2} \tau^2 \sum_{j,j'=0}^{\kappa-1} \tau^{j+j'} \beta_k^{(j)} \beta_k^{(j')} \lambda_k^2 + \ldots \right). \tag{17}$$

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[4] There is a misprint in equation (6) of [14] which is corrected here.

Collecting powers of τ we find

$$-\mathrm{i}\,\tau\sum_{k=1}^{n^2-1}\alpha_k^{(0)}\,\lambda_k - \mathrm{i}\,\tau^2\sum_{k=1}^{n^2-1}\alpha_k^{(1)}\,\lambda_k - \tfrac{1}{2}\,\tau^2\sum_{k=1}^{n^2-1}\left(\alpha_k^{(0)}\right)^2\lambda_k^2 - \tfrac{1}{2}\,\tau^2\sum_{k\neq k'}^{n^2-1}\alpha_k^{(0)}\,\alpha_{k'}^{(0)}\,\lambda_k\,\lambda_{k'} + \ldots$$

$$= \prod_{k=1}^{n^2-1}\left(1 - \mathrm{i}\,\tau\,\beta_k^{(0)}\,\lambda_k - \mathrm{i}\,\tau^2\,\beta_k^{(1)}\,\lambda_k - \tfrac{1}{2}\,\tau^2\left(\beta_k^{(0)}\right)^2\lambda_k^2 + \ldots\right) - 1$$

$$= -\mathrm{i}\,\tau\sum_{k=1}^{n^2-1}\beta_k^{(0)}\,\lambda_k - \mathrm{i}\,\tau^2\sum_{k=1}^{n^2-1}\beta_k^{(1)}\,\lambda_k - \tfrac{1}{2}\,\tau^2\sum_{k=1}^{n^2-1}\left(\beta_k^{(0)}\right)^2\lambda_k^2$$

$$-\tau^2\sum_{k'>k}^{n^2-1}\beta_k^{(0)}\,\beta_{k'}^{(0)}\,\lambda_k\,\lambda_{k'} + \ldots. \tag{18}$$

Multiplying (18) by λ_p and using (12) yields

$$\tau\,\alpha_p^{(0)} + \tau^2\alpha_p^{(1)} - \frac{\mathrm{i}}{4}\,\tau^2\sum_{k'>k}^{n^2-1}\alpha_k^{(0)}\alpha_{k'}^{(0)}\,\mathrm{tr}\,\lambda_p(\lambda_k\,\lambda_{k'} + \lambda_{k'}\,\lambda_k) + \ldots$$

$$= \tau\,\beta_p^{(0)} + \tau^2\beta_p^{(1)} - \frac{\mathrm{i}}{2}\,\tau^2\sum_{k'>k}^{n^2-1}\beta_k^{(0)}\,\beta_{k'}^{(0)}\,\mathrm{tr}\,\lambda_p\,\lambda_k\,\lambda_{k'} + \ldots. \tag{19}$$

Equating like powers of τ we have

$$\beta_p^{(0)} = \alpha_p^{(0)} \tag{20}$$

$$\beta_p^{(1)} = \alpha_p^{(1)} + \frac{\mathrm{i}}{4}\sum_{k'>k}^{n^2-1}\alpha_k^{(0)}\,\alpha_{k'}^{(0)}\,\mathrm{tr}\,\lambda_p\,[\lambda_k,\lambda_{k'}] \tag{21}$$

$$\vdots$$

Clearly, one can continue this expansion to obtain equations for $\beta_p^{(2)}, \ldots, \beta_p^{(\kappa-1)}$. Moreover this technique is well suited to straightforward implementation in a symbolic algebra language. In such an implementation, one uses the explicit form of the λ_k to obtain a matrix equation at each order, which naturally leads to a set of linear equations for $\beta_k^{(j)}$. This approach eliminates the need for developing explicit expressions for $\beta_k^{(j)}$.

4. An example

By way of example, we consider a two-level atom with population-preserving decay [11]. The Hamiltonian for this system is

$$H = \begin{pmatrix} \epsilon & \Omega(t) \\ \Omega(t) & -\epsilon \end{pmatrix} \tag{22}$$

where, for simplicity, we have taken $\Omega(t)$ to be real. The dissipation operator is given by

$$\Lambda[\rho] = \begin{pmatrix} \gamma\,\rho_{22} & -\tfrac{1}{2}\,\gamma\,\rho_{12} \\ -\tfrac{1}{2}\,\gamma\,\rho_{21} & -\gamma\,\rho_{22} \end{pmatrix}. \tag{23}$$

As we explained above, we are interested in the case of weak dissipation, i.e., in $\gamma \ll \epsilon$.

Following the notation of (13), for a second-order integrator we have $\alpha_1 = \Omega + \tfrac{1}{2}\,\tau\,\Omega'$, $\alpha_2 = 0$ and $\alpha_3 = \epsilon$, and we need to compute β_k through order τ. From (20) and (21) we

find $\beta_1 = \Omega + \frac{1}{2} \tau \Omega'$, $\beta_2 = \tau \epsilon \Omega$, and $\beta_3 = \epsilon$. Explicitly the unitary integrator is

$$
\mathcal{U} = \begin{bmatrix} \cos(\Omega \tau + \frac{1}{2} \Omega' \tau^2) & -\mathrm{i} \sin(\Omega \tau + \frac{1}{2} \Omega' \tau^2) \\ -\mathrm{i} \sin(\Omega \tau + \frac{1}{2} \Omega' \tau^2) & \cos(\Omega \tau + \frac{1}{2} \Omega' \tau^2) \end{bmatrix}
$$
$$
\times \begin{bmatrix} \cos(\epsilon \Omega \tau^2) & -\sin(\epsilon \Omega \tau^2) \\ \sin(\epsilon \Omega \tau^2) & \cos(\epsilon \Omega \tau^2) \end{bmatrix} \begin{bmatrix} \mathrm{e}^{-\mathrm{i}\epsilon\tau} & 0 \\ 0 & \mathrm{e}^{\mathrm{i}\epsilon\tau} \end{bmatrix}. \tag{24}
$$

We obtain an evolution operator, $\mathcal{G}(\tau)$, for the dissipative part by solving

$$
\dot{\rho} = \Lambda[\rho] \tag{25}
$$

using the midpoint rule

$$
\varrho_{n+1} - \varrho_n = \tau \Lambda[\tfrac{1}{2}(\varrho_{n+1} + \varrho_n)]. \tag{26}
$$

We can interpret (26) as differencing (25) at $t + \frac{1}{2}\tau$, which yields an algorithm accurate to second order. Since Λ is a linear operator, (26) can be solved directly to provide an explicit formula for ϱ_{n+1}:

$$
\varrho_{n+1} = \mathcal{G}(\tau)\varrho_n
$$
$$
= \begin{bmatrix} (\varrho_n)_{11} + \dfrac{\tau \gamma}{1 + \frac{1}{2}\tau\gamma}(\varrho_n)_{22} & \dfrac{1 - \frac{1}{4}\tau\gamma}{1 + \frac{1}{4}\tau\gamma}(\varrho_n)_{12} \\[3mm] \dfrac{1 - \frac{1}{4}\tau\gamma}{1 + \frac{1}{4}\tau\gamma}(\varrho_n)_{21} & \dfrac{1 - \frac{1}{2}\tau\gamma}{1 + \frac{1}{2}\tau\gamma}(\varrho_n)_{22} \end{bmatrix}. \tag{27}
$$

Following (2), we can write the total evolution operator as

$$
\varrho_{n+1} = \mathcal{G}(\tfrac{1}{2}\tau) \circ \mathcal{U}(t,\tau) \circ \mathcal{G}(\tfrac{1}{2}\tau)\,\varrho_n \tag{28}
$$

where

$$
\mathcal{U}(t,\tau)\,\varrho_n = U\varrho_n U^\dagger \tag{29}
$$

is the unitary integrator. Note that both $\mathrm{tr}\,\mathcal{G} = 1$ and $\mathrm{tr}\,\mathcal{U} = 1$ and hence $\mathrm{tr}\,\varrho_{n+1} = \mathrm{tr}\,\varrho_n$.

Below we compare the results of using this split-operator method with the results of a second-order predictor–corrector algorithm (Heun's method [20]). Applying the predictor–corrector formulae to (3) gives

$$
\varrho_{n+1} = \varrho_n - \mathrm{i}\tau\left[H(n\tau) + H((n+1)\tau), \varrho_n\right] + \tau\Lambda[\varrho_n] + \tfrac{1}{2}\tau^2\Lambda\left[\Lambda[\varrho_n]\right]
$$
$$
- \tfrac{1}{2}\tau^2\left[H((n+1)\tau), \left[H(n\tau), \varrho_n\right]\right] - \tfrac{1}{2}\mathrm{i}\tau^2\Lambda\left[\left[H((n+1)\tau), \varrho_n\right]\right]
$$
$$
- \tfrac{1}{2}\mathrm{i}\tau^2\left[H((n+1)\tau), \Lambda[\varrho_n]\right] \tag{30}
$$

where we have made use of the fact that Λ is linear. Now, as expected, $\mathrm{tr}\,\varrho_{n+1} - \mathrm{tr}\,\varrho_n = 0$, since the trace of a commutator vanishes as does $\mathrm{tr}\,\Lambda$, thus this algorithm also exactly conserves the population.

We provide a pair of numerical examples to illustrate the advantage of the structure-preserving approach over the general-purpose algorithm. We take the interaction to be a Gaussian pulse

$$
\Omega(t) = \Omega_0\,\mathrm{e}^{-(t-t_0)^2/\sigma^2} \tag{31}
$$

and use the parameters

$$
\epsilon = 1, \quad \Omega_0 = \tfrac{1}{2}, \quad t_0 = 10, \quad \sigma = \tfrac{20}{3} \text{ and } \gamma = \tfrac{1}{20}. \tag{32}
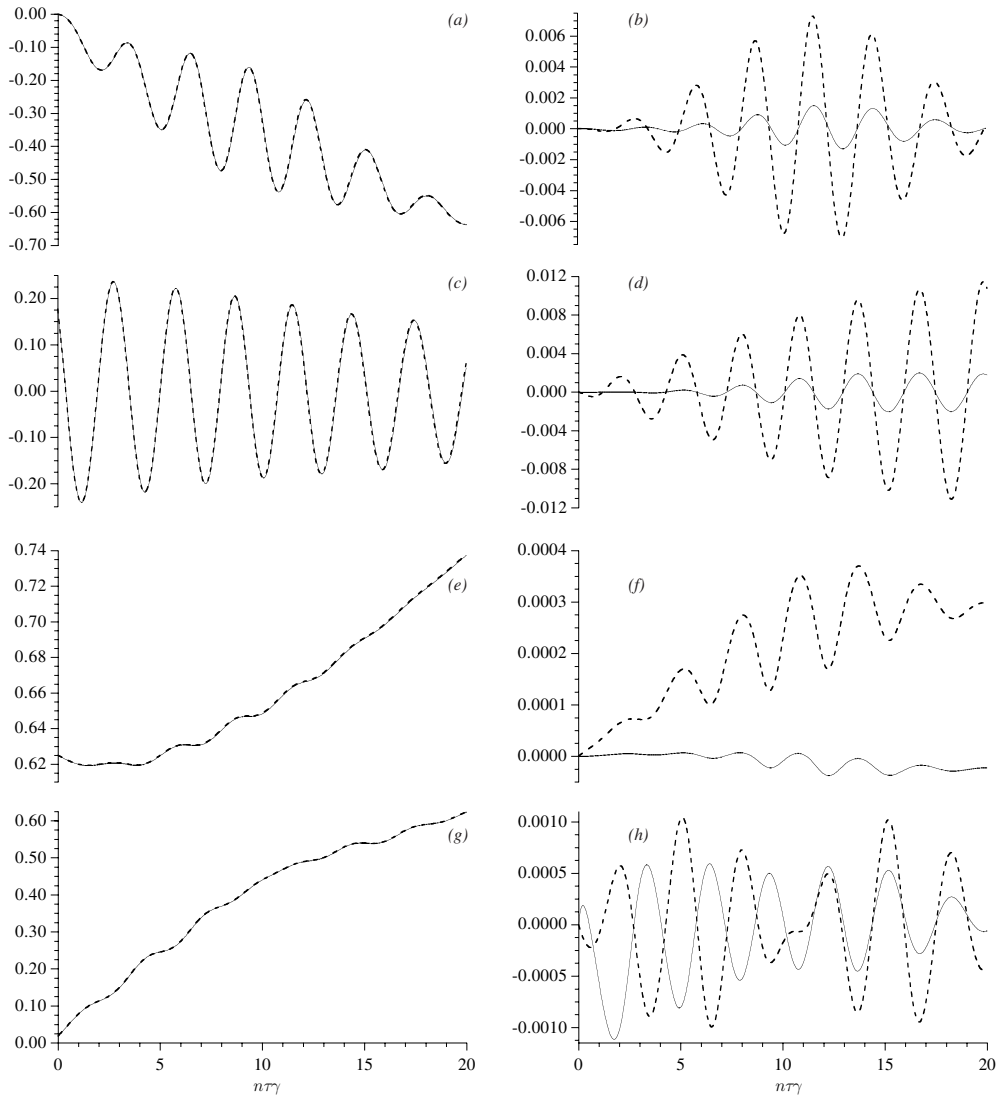$$

Figure 1. The density matrix from a numerical solution of (3) with the Hamiltonian given by (22) and (31), the dissipation is given by (23), where $\epsilon = 1, \Omega_0 = \frac{1}{2}, t_0 = 10, \sigma = \frac{20}{3}, \gamma = \frac{1}{20}, \tau = \frac{1}{20}$. The initial condition is given by (33). The heavy dashes and solid lines denote the predictor–corrector and unitary integrator solutions, respectively. The panels are: inversion $((\varrho_n)_{22} - (\varrho_n)_{11})$ (a); error in the inversion (b); Im $(\varrho_n)_{12}$ (c); error in Im $(\varrho_n)_{12}$ (d); tr ϱ_n^2 (e); error in tr ϱ_n^2 (f); energy expectation value (tr $H\varrho_n$) (g); and energy error (h).

We set the initial condition

$$\rho(t = 0) = \begin{bmatrix} \frac{1}{2} & \frac{1}{4}\,\mathrm{e}^{\mathrm{i}\pi/4} \\ \frac{1}{4}\,\mathrm{e}^{-\mathrm{i}\pi/4} & \frac{1}{2} \end{bmatrix} \tag{33}$$

and use the same time step $\tau = \frac{1}{20}$ for both methods. The first set of numerical results is shown in figure 1. Clearly the unitary integrator produces smaller errors in the inversion and coherence elements, and is also somewhat better in the energy than predictor–corrector. As
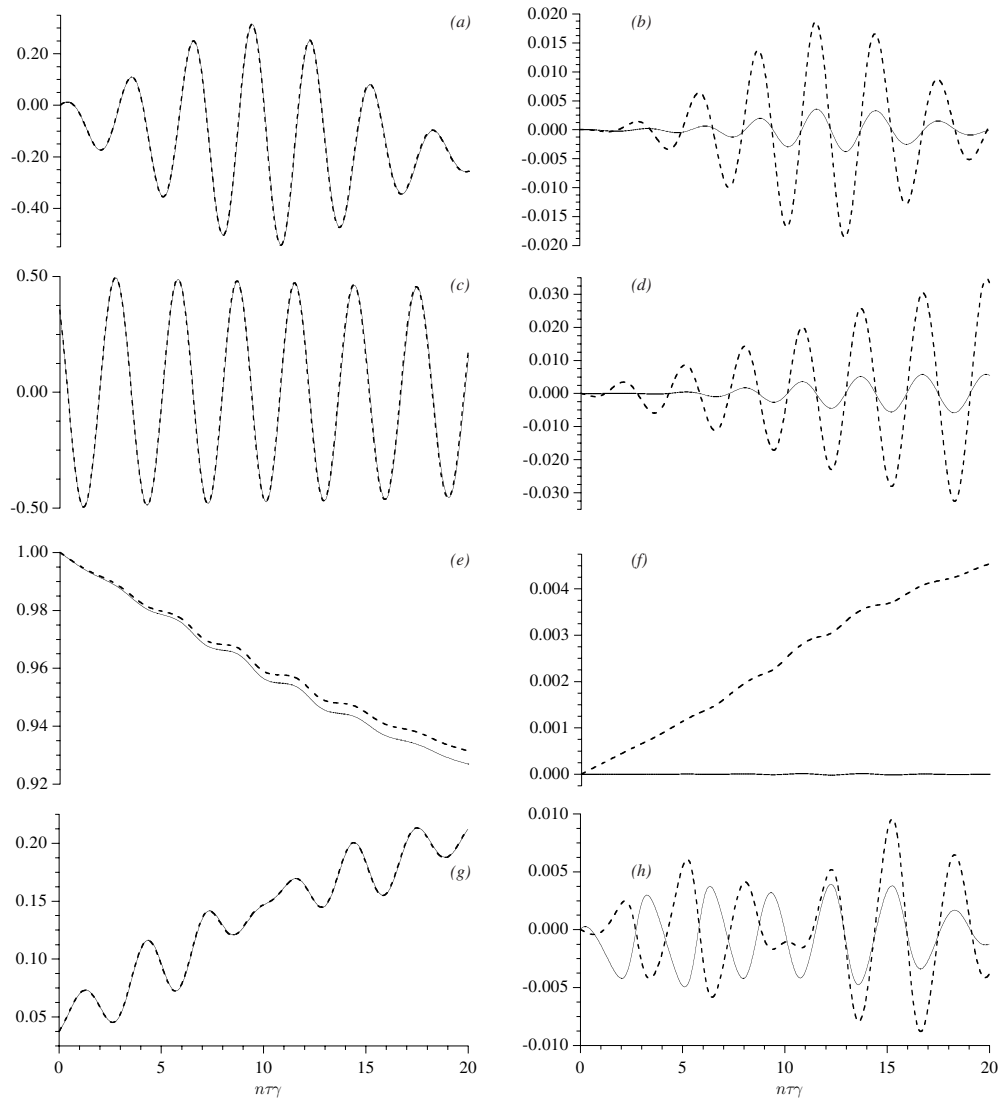
Figure 2. The density matrix as in figure 1 but with $\gamma = \frac{1}{100}$.

with symplectic integrators, the nature of the energy error is not surprising. Since the unitary integrator is effectively constructed from an approximate Hamiltonian, we cannot expect tr $H\varrho_n$ to be computed exactly but rather we expect the error in tr $H\varrho_n$ to oscillate rather than exhibit secular growth [14].

Figure 2 shows numerical results with the same parameters as above except for $\gamma = 1/100$. In both cases, the energy error for both methods is quite similar and provides little insight into which method provides the more accurate solution. The errors in tr ρ^2 are more telling: the operator-splitting method clearly performs better. In figure 2(e), we see how the structural errors introduced by the predictor–corrector algorithm reduce the decoherence caused by the dissipation.

4.1. Discussion

In view of the results shown, we considered several different approaches to handling the evolution of the dissipative operator, in order to understand the origin of the superior performance of the algorithm based on the unitary integrator. In addition to using the midpoint rule, splittings based on a predictor–corrector solution to (25) as well as splittings based on the exact solution of (25) were explored. (Since the exact solution requires the evaluation of an exponential, it is much more efficient to solve the differential equation than to employ the exact solution.) In each of these cases the full evolution was computed via (28) with the appropriate choice for $\mathcal{G}$. Regardless of the method of handling the dissipation, essentially the same results were obtained; clearly the advantage of the operator-splitting approach is due to the use of the unitary integrator.

To understand these results better, it is worthwhile to examine the numerical methods in detail through backwards error analysis [3–7]. In section 1, we argued that dissipation operators are typically sufficiently simple to not possess a 'structure' that is physically important. This assertion is borne out by the true dynamical equations corresponding to the various numerical solutions. For the full system (3), this analysis is rather cumbersome, so we consider separately the Hamiltonian and dissipative terms.

We begin by examining the consequence of applying the midpoint rule to

$$\dot{\rho} = \Lambda_{\text{eff}}[\rho] \equiv \begin{bmatrix} \gamma_1\,\rho_{22} & -\frac{1}{2}\,\gamma_2\,\rho_{12} \\ -\frac{1}{2}\,\gamma_2\,\rho_{21} & -\gamma_1\,\rho_{22} \end{bmatrix}. \tag{34}$$

The idea is to determine γ_1 and γ_2 such that when the midpoint rule is applied to (34) we obtain the *exact* solution of (25). Since the midpoint rule is second order, we expect $\gamma_{1,2} = \gamma + \mathcal{O}(\tau^2)$. The procedure is to compare the midpoint rule solution of (34) with the exact solution of (25), generated from the Taylor series expansion about $\tau = 0$. Some straightforward algebra gives

$$\gamma_1 = \gamma - \tfrac{1}{12}\,\tau^2\gamma^3 + \tfrac{1}{120}\,\tau^4\gamma^5 + \mathcal{O}(\tau^6\gamma^7) \tag{35}$$

$$\gamma_2 = \gamma - \tfrac{1}{48}\,\tau^2\gamma^3 + \tfrac{1}{1920}\,\tau^4\gamma^5 + \mathcal{O}(\tau^6\gamma^7). \tag{36}$$

What this says is that when we use the midpoint rule to solve (25) we are in fact getting the exact solution of the slightly modified system (34). Now although $\gamma_1 \neq \gamma_2$, the physical meaning of the dissipation operator is only slightly changed since

$$\frac{\gamma_1}{\gamma_2} = 1 - \tfrac{1}{16}\,\tau^2\gamma^2 + \mathcal{O}(\tau^4\gamma^4). \tag{37}$$

The important point is that this modified dissipation operator is physically reasonable, with qualitative and quantitative behaviour very similar to the original dissipation mechanism. This is precisely what it means not to have a delicate structure; the quantitative and qualitative behaviour is robust to small perturbations in the form of the dynamical equations. However, had the dissipation model been more complex, it is quite likely that to determine Λ_{eff} it would have been necessary to introduce new couplings in the equations which would most likely lead to qualitatively different physical content. This is exactly what we will see below when we perform a similar analysis on the Hamiltonian part of (3).

Performing this same analysis for the predictor–corrector solution of (25) yields a very similar result. In this case we find

$$\gamma_1 = \gamma + \tfrac{1}{6}\,\tau^2\,\gamma^3 + \tfrac{1}{8}\,\tau^3\,\gamma^4 + \tfrac{2}{15}\,\tau^4\,\gamma^5 + \mathcal{O}(\tau^5\gamma^6) \tag{38}$$

$$\gamma_2 = \gamma + \tfrac{1}{24}\,\tau^2\,\gamma^3 + \tfrac{1}{64}\,\tau^3\,\gamma^4 + \tfrac{1}{120}\,\tau^4\,\gamma^5 + \mathcal{O}(\tau^5\gamma^6) \tag{39}$$

and

$$\frac{\gamma_1}{\gamma_2} = 1 - \tfrac{1}{8}\,\tau^2\gamma^2 + \mathcal{O}(\tau^3\gamma^3) \tag{40}$$

leading us to the same basic conclusion as for the midpoint algorithm.

We now turn to an analysis of the numerical solution of the purely Hamiltonian system (4). For this system, the predictor–corrector time advance is

$$\varrho_{n+1} = \varrho_n - \mathrm{i}\,\tau\left[H(n\tau) + H((n+1)\tau), \varrho_n\right] - \tfrac{1}{2}\,\tau^2\left[H((n+1)\tau), \left[H(n\tau), \varrho_n\right]\right]. \tag{41}$$

In the same fashion as above, we seek to determine the effective Hamiltonian

$$H_{\mathrm{eff}} = \begin{bmatrix} \epsilon_{\mathrm{eff}} & \Omega_{\mathrm{eff}} \\ \Omega_{\mathrm{eff}}^* & -\epsilon_{\mathrm{eff}} \end{bmatrix}. \tag{42}$$

such that (41) applied using H_{eff} gives the exact solution to (4). It turns out that for predictor–corrector, it is possible to achieve this goal only through third order in τ; the fourth-order corrections result in a non-hermitian H_{eff}. To third order we find:

$$\epsilon_{\mathrm{eff}} = \epsilon\left[1 - \tfrac{2}{3}\,\tau^2\left(\Omega^2 + \epsilon^2\right)\right] \tag{43}$$

$$\Omega_{\mathrm{eff}} = \Omega\left[1 - \tfrac{2}{3}\,\tau^2\left(\Omega^2 + \epsilon^2\right)\right] - \tfrac{1}{12}\,\tau^2\,\Omega'' - \tfrac{1}{3}\,\mathrm{i}\,\tau^2\,\Omega'. \tag{44}$$

At fourth order, the effective system is no longer Hamiltonian but contains dissipation, thus we expect the predictor–corrector to introduce errors in the Hioe–Eberly invariants at fourth order (for example, see equation (11) in [14]). This calculation becomes increasingly complex; we can adequately illustrate our point more clearly by considering a constant Hamiltonian. At fourth order, solving (4) with (41) is equivalent to the exact solution of

$$\hbar\,\dot\rho = -\mathrm{i}\,[H_{\mathrm{eff}}, \rho] + \Lambda_{\mathrm{eff}}[\rho] \tag{45}$$

with

$$\epsilon_{\mathrm{eff}} = \epsilon\left[1 - \tfrac{2}{3}\,\tau^2\left(\Omega^2 + \epsilon^2\right)\right] \tag{46}$$

$$\Omega_{\mathrm{eff}} = \Omega\left[1 - \tfrac{2}{3}\,\tau^2\left(\Omega^2 + \epsilon^2\right)\right] + \mathrm{i}\,\tau^3\epsilon\,\Omega\left(\Omega^2 + \epsilon^2\right) \tag{47}$$

and

$$\begin{aligned}
\Lambda_{\mathrm{eff}}[\rho] = \tau^3\,\Omega^2\left(\Omega^2 + \epsilon^2\right) &\begin{bmatrix} \rho_{22} - \rho_{11} & \rho_{21} - \rho_{12} \\ \rho_{12} - \rho_{21} & \rho_{11} - \rho_{22} \end{bmatrix} \\
+ 2\,\tau^3\,\epsilon\,\Omega\left(\Omega^2 + \epsilon^2\right) &\begin{bmatrix} 0 & \rho_{11} - \rho_{22} \\ \rho_{11} - \rho_{22} & 0 \end{bmatrix} \\
- 2\,\tau^3\,\epsilon^2\left(\Omega^2 + \epsilon^2\right) &\begin{bmatrix} 0 & \rho_{12} \\ \rho_{21} & 0 \end{bmatrix}.
\end{aligned} \tag{48}$$

Notice that the form of Λ_{eff} describes a much different set of physical processes than those responsible for the actual dissipation in (3). For example, the presence of the inversion in the off-diagonal elements and the fact that Λ_{eff} drives the system towards equal populations rather than decay to the lower state. Although $\|\Lambda_{\mathrm{eff}}\| \ll \|\Lambda\|$, the numerical results show that Λ_{eff} is a sufficient perturbation to significantly affect the solution. Thus even though (3) does not exactly preserve the c_j, the remnants of the ideal structure of (4) are sufficiently strong that the perturbations introduced by the predictor–corrector are enough to adversely affect the accuracy of the numerical solution.

5. Conclusions

The advantages of unitary integration are seen most dramatically in ideal or nearly ideal systems sensitive to small errors. The important point to be made here, however, is that the operator-splitting approach where a unitary integrator is used for the Hamiltonian evolution is

guaranteed to recover the ideal solution in the limit of small dissipation, whereas this is not the case for a generic method. This is an important consideration when studying near-ideal situations where decoherence is small, but may be the primary point of interest or limiting factor (as in studies of quantum computation in real systems, for example). Examples of such systems include (i) those where the time scale of interest is very long compared to the natural scales of the problem, (ii) systems with sensitive dependence upon initial conditions and (iii) systems where dissipation and decoherence are small, but of significant importance. Further areas of future investigation include application of these methods to such systems as (i) atomic clocks of various types [21], and (ii) numerical studies of decoherence in quantum computers. This latter application may include studies of decoherence-free subspaces, for which the structure of the dissipation operator is of significant importance.

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