On Some Difficulties in Integrating Highly Oscillatory Hamiltonian Systems

Uri M. Ascher[1]* and Sebastian Reich[2]

[1] Institute of Applied Mathematics and Department of Computer Science, University of British Columbia, Vancouver, B.C., Canada V6T 1Z4 (ascher@cs.ubc.ca)

[2] Konrad-Zuse-Zentrum, Takustr. 7, D-14195 Berlin, Germany (reich@zib.de)

Abstract. The numerical integration of highly oscillatory Hamiltonian systems, such as those arising in molecular dynamics or Hamiltonian partial differential equations, is a challenging task. Various methods have been suggested to overcome the step-size restrictions of explicit methods such as the Verlet method. Among these are multiple-time-stepping, constrained dynamics, and implicit methods. In this paper, we investigate the suitability of time-reversible, semi-implicit methods. Here semi-implicit means that only the highly oscillatory part is integrated by an implicit method such as the midpoint method or an energy-conserving variant of it. The hope is that such methods will allow one to use a step-size k which is much larger than the period ε of the fast oscillations.

However, our results are not encouraging. Even in the absence of resonance-type instabilities, we show that in general one must require that k^2/ε be small enough. Otherwise the method might become unstable and/or it might lead to a wrong approximation of the slowly varying solution components. The latter situation might, in some cases, even require that k/ε be small in order to avoid this danger. While certain (semi-implicit) energy conserving methods prove to be robust for some model problems, they may also yield deceptively-looking, wrong solutions for other simple model problems, in circumstances where the corresponding constrained dynamics formulation may not be easily derived and used.

1 Introduction

In this paper, we discuss semi-implicit/implicit integration methods for highly oscillatory Hamiltonian systems. Such systems arise, for example, in molecular dynamics [1] and in the finite dimensional truncation of Hamiltonian partial differential equations. Classical discretization methods, such as the Verlet method [19], require step-sizes k smaller than the period ε of the fast oscillations. Then these methods find pointwise accurate approximate solutions. But the time-step restriction implies an enormous computational burden. Furthermore, in many cases the high-frequency responses are of little or no interest. Consequently, various researchers have considered the use of semi-implicit/implicit methods, e.g. [6,11,9,16,18,12,13,8,17,3].

* The work of this author was partially supported under NSERC Canada Grant OGP0004306.

A popular implicit discretization is the (implicit) midpoint method [7] which, applied to a system of the type

$$\frac{d}{dt}\boldsymbol{q} = \boldsymbol{p}\,, \tag{1a}$$
$$\frac{d}{dt}\boldsymbol{p} = -\boldsymbol{\nabla} U(\boldsymbol{q})\,, \tag{1b}$$

yields the discretization

$$\boldsymbol{q}_{n+1} = \boldsymbol{q}_n + k\,\boldsymbol{p}_{n+1/2}\,, \tag{2a}$$
$$\boldsymbol{p}_{n+1} = \boldsymbol{p}_n - k\,\boldsymbol{\nabla} U(\boldsymbol{q}_{n+1/2}) \tag{2b}$$

with $\boldsymbol{p}_{n+1/2} = [\boldsymbol{p}_{n+1} + \boldsymbol{p}_n]/2$, etc. The behavior of the midpoint method when applied to highly-oscillatory systems

$$\frac{d}{dt}\boldsymbol{q} = \boldsymbol{p}\,, \tag{3a}$$
$$\frac{d}{dt}\boldsymbol{p} = -\boldsymbol{\nabla} W(\boldsymbol{q}) - \kappa g(\boldsymbol{q})\boldsymbol{\nabla} g(\boldsymbol{q})\,, \tag{3b}$$

W, g potential functions, $\kappa \gg 1$, has been discussed in various papers (see, for example, [6,11,9,16,3]). It has been pointed out that, for step-sizes $k \gg \varepsilon := 1/\sqrt{\kappa}$, the midpoint method can become unstable due to resonances [9,16], i.e., for specific values of k. However, generic instabilities arise if the step-size k is chosen such that k^2/ε is not small [3,6,18]. For systems with a rotational symmetry this has been shown rigorously in [6]. This effect is generic for highly oscillatory Hamiltonian systems, as argued for in [3] in terms of decoupling transformations and proved for a linear time varying system without symmetry.

Even further complications are to be expected for general systems of the type (3). These are related to the approximation of the slowly varying solution components and other related quantities of (3) for $\kappa \to \infty$ by the corresponding solution of the constrained system DAE

$$\begin{aligned} \frac{d}{dt}\boldsymbol{q} &= \boldsymbol{p}\,, \\ \frac{d}{dt}\boldsymbol{p} &= -\boldsymbol{\nabla} W(\boldsymbol{q}) - \lambda\boldsymbol{\nabla} g(\boldsymbol{q})\,, \\ 0 &= g(\boldsymbol{q})\,. \end{aligned} \tag{4}$$

In general, the solution components of the DAE (4) are the correct limits (as $\kappa \to \infty$) of the corresponding slowly varying solution components of the free dynamics only if an additional (conservative) force term is introduced in the constrained system [14,5]. It turns out [3] that the midpoint method may falsely approximate this correcting force term to zero unless $k = \mathcal{O}(\varepsilon)$, which leads to a step-size restriction of the same order of magnitude as explicit methods such as the Verlet method! In cases where the additional force term is zero this further complication does not apply, of course. The specific form of the additional force term depends on the initial conditions and the potential function g in (3a)-(3b).

Since fully implicit methods are very expensive when long-range forces are present, we consider semi-implicit methods that are based on the splitting of the force field into weak forces and strong forces, the latter causing the high-frequency oscillations [12]. The limitations of the methods considered here are not related to this splitting, i.e., corresponding fully implicit methods would not remove these limitations.

The purpose of this paper is twofold: (i) We summarize possible difficulties with the midpoint method (other than resonance instability, which has been treated extensively elsewhere) by looking at a simple (molecular) model problem. (ii) We investigate the suitability of some energy conserving methods.

Regarding the second objective (ii), it has been argued [6] that energy conserving methods might be better suited for the numerical integration of highly oscillatory problems such as (3). We show that such a statement must be made with extreme caution. The main concern is that energy conserving methods might wrongly approximate slowly varying solution quantities. Even worse, unlike the midpoint method where a blatant non-conservation of energy indicates potential trouble, energy conserving methods, by definition, do not provide such a warning flag. Thus, energy conserving methods are potentially more misleading on one hand, and they do not seem to provide a significant gain on the other hand. Consequently, we generally discourage their use for highly oscillatory Hamiltonian systems, unless special circumstances warrant it.

2 Some Energy Conserving Methods

Given a general autonomous, separable Hamiltonian system (1), the Hamiltonian

$$H(\mathbf{q},\mathbf{p}) = \frac{1}{2}\mathbf{p}^T\mathbf{p} + U(\mathbf{q})$$

remains constant, of course, along exact solution trajectories. Here we wish an approximate, numerical solution to preserve this invariant as well, i.e.,

$$H(\mathbf{q}_{n+1},\mathbf{p}_{n+1}) = H(\mathbf{q}_n,\mathbf{p}_n), \quad n = 0, 1, \ldots . \tag{5}$$

Moreover, we want the resulting numerical method to remain time-reversible, which precludes the most obvious projection schemes from further consideration.

Time-reversible energy conserving methods can be obtained by appropriate modifications to the (time-reversible) midpoint method. Two such modifications are: (i) scaling of the force field by a scalar such that total energy is conserved [6,12], and (ii) using a symmetric projection technique onto the hypersurface of constant energy. Here are the details.

2.1 The Simo-Gonzales Method

The method considered in [6] can be derived as follows. Let us assume that $U(\mathbf{q}) = V(r)$, where $r = \|\mathbf{q}\|$. Then (1b) reads

$$\frac{d}{dt}\boldsymbol{p} = -\frac{V'(r)}{r}\boldsymbol{q}\,.$$

Consider for the nth time step the linear harmonic oscillator

$$\bar{H}(\mathbf{q},\mathbf{p}) = \frac{1}{2}\mathbf{p}^T\mathbf{p} + \frac{\sigma}{2}\mathbf{q}^T\mathbf{q}$$

where $\sigma = \sigma_n$ is constant. Discretizing the equations of motion using the midpoint method

$$\mathbf{q}_{n+1} = \mathbf{q}_n + k\mathbf{p}_{n+1/2} \tag{6a}$$

$$\mathbf{p}_{n+1} = \mathbf{p}_n - k\sigma\mathbf{q}_{n+1/2} \tag{6b}$$

yields $\bar{H}(\mathbf{q}_{n+1},\mathbf{p}_{n+1}) = \bar{H}(\mathbf{q}_n,\mathbf{p}_n)$, because $\bar{H}$ is quadratic [2]. Comparing this to (5) we see that (5) is satisfied if we choose

$$\sigma = 2\frac{V(r_{n+1}) - V(r_n)}{r_{n+1}^2 - r_n^2} = \frac{2}{r_{n+1} + r_n}\,\frac{V(r_{n+1}) - V(r_n)}{r_{n+1} - r_n}\,, \tag{6c}$$

where $r_n = \|\mathbf{q}_n\|$. The method (6) is then energy conserving. It also conserves linear and angular momentum.

The energy conserving method (6) is a close variation of the method

$$\mathbf{q}_{n+1} = \mathbf{q}_n + k\mathbf{p}_{n+1/2} \tag{7a}$$

$$\mathbf{p}_{n+1} = \mathbf{p}_n - k\frac{V'(r_{n+1/2})}{r_{n+1/2}}\mathbf{q}_{n+1/2} \tag{7b}$$

(because $\frac{V(r_{n+1})-V(r_n)}{r_{n+1}-r_n}$ can be viewed as a difference approximation of $V'(r_{n+1/2})$) where

$$r_{n+1/2} = \frac{1}{2}(r_{n+1} + r_n) = \frac{1}{2}(\|\mathbf{q}_{n+1}\| + \|\mathbf{q}_n\|)\,. \tag{7c}$$

Note that this latter method differs from the midpoint method, where one would use $r(\mathbf{q}_{n+1/2}) = \|(\mathbf{q}_{n+1} + \mathbf{q}_n)/2\|$ instead of (7c) for $r_{n+1/2}$ in (7b). For highly oscillatory systems with $k \gg \varepsilon$, this can be a significant difference, because r is discretized directly in (7). An example in §4 below shows that the midpoint method can become unstable while (7) and (6) remain stable.

For a general Hamiltonian system (1), a straightforward generalization of (6) reads

$$\mathbf{q}_{n+1} = \mathbf{q}_n + k\mathbf{p}_{n+1/2} \tag{8a}$$

$$\mathbf{p}_{n+1} = \mathbf{p}_n - k\sigma\boldsymbol{\nabla}U(\boldsymbol{q}_{n+1/2}) \tag{8b}$$

where

$$\sigma = \frac{U(\mathbf{q}_{n+1}) - U(\mathbf{q}_n)}{\boldsymbol{\nabla}U(\boldsymbol{q}_{n+1/2})^T(\mathbf{q}_{n+1} - \mathbf{q}_n)}\,. \tag{8c}$$

For refined variants of this method in the context of many-particle systems, see [15,12].

2.2 A Projection Method

Here a symmetric projection step is used to enforce conservation of energy. Let $\boldsymbol{a}(\boldsymbol{q},\boldsymbol{p})$ and $\boldsymbol{b}(\boldsymbol{q},\boldsymbol{p})$ be two vector-valued functions such that $(\boldsymbol{p}^T\boldsymbol{a}(\boldsymbol{q},\boldsymbol{p}) + \boldsymbol{\nabla}U(\boldsymbol{q})^T\boldsymbol{b}(\boldsymbol{q},\boldsymbol{p}))$ is bounded away from zero. Then we propose the following modified midpoint method,

$$\begin{aligned}
\boldsymbol{q}_{n+1} &= \boldsymbol{q}_n + k\,\boldsymbol{p}_{n+1/2} + \lambda\,\boldsymbol{b}(\boldsymbol{q}_{n+1/2},\boldsymbol{p}_{n+1/2})\,,\\
\boldsymbol{p}_{n+1} &= \boldsymbol{p}_n - k\,\boldsymbol{\nabla}U(\boldsymbol{q}_{n+1/2}) + \lambda\,\boldsymbol{a}(\boldsymbol{p}_{n+1/2},\boldsymbol{p}_{n+1/2})\,,\\
0 &= H(\boldsymbol{q}_{n+1},\boldsymbol{p}_{n+1}) - H(\boldsymbol{q}_n,\boldsymbol{p}_n)\,.
\end{aligned}$$

The parameter λ is determined by the requirement that the total energy $H = \boldsymbol{p}^T\boldsymbol{p}/2 + U(\boldsymbol{q})$ is conserved.

The particular choice $\boldsymbol{a}(\boldsymbol{q},\boldsymbol{p}) = \boldsymbol{\nabla}U(\boldsymbol{q})$ and $\boldsymbol{b}(\boldsymbol{q},\boldsymbol{p}) = 0$ leads to the method (8). This is further discussed in [12].

Another option is $\boldsymbol{a}(\boldsymbol{q},\boldsymbol{p}) = \boldsymbol{p}$ and $\boldsymbol{b}(\boldsymbol{q},\boldsymbol{p}) = \boldsymbol{\nabla}U(\boldsymbol{q})$. This guarantees that we are discretizing a pure index-2 DAE for which λ is well-defined. But for this choice we observed severe difficulties with Newton's method, where a step-size smaller even than what is required by explicit methods is needed to obtain convergence. In fact, it can be shown that when the linear harmonic oscillator is cast into such a projected DAE, the linearized problem can easily become unstable for $k > \varepsilon$. Another way is to check the conditions of the Newton-Kantorovich Theorem, which guarantees convergence of the Newton method. These conditions are also found to be satisfied only for a very small step size k, if ε is small.

This latter modified midpoint method does work well, however, for the long time integration of Hamiltonian systems which are not highly oscillatory. Note that conservation of any other first integral can be enforced in a similar manner. To our knowledge, this method has not been considered in the literature before in the context of Hamiltonian systems, although it is standard among methods for incompressible Navier-Stokes (where its time-reversibility is not an issue, however).

For highly oscillatory Hamiltonian systems, the best energy conserving midpoint variant that we are aware of is (6). In the sequel we therefore examine only its performance.

3 A Simple Model Problem

As our first model problem, we take the motion of a diatomic molecule under an external force field. For simplicity, it is assumed that (i) the motion is planar, (ii) the two atoms have equal mass $m = 1$, and (iii) the chemical bond is modeled by a stiff harmonic spring with equilibrium length $r_0 = 1$. Denoting the positions of the two atoms by $\boldsymbol{q}_i \in \mathcal{R}^2$, $i = 1, 2$, the corresponding Hamiltonian function is of type

$$H = \frac{\boldsymbol{p}_1^T \boldsymbol{p}_1}{2} + \frac{\boldsymbol{p}_2^T \boldsymbol{p}_2}{2} + \frac{\kappa}{2}(\|\boldsymbol{q}_1 - \boldsymbol{q}_2\| - 1)^2 + V_1(\|\boldsymbol{q}_1 - \boldsymbol{q}_1^0\|) + V_2(\|\boldsymbol{q}_2 - \boldsymbol{q}_2^0\|) \,.$$

Here $\kappa \gg 1$ is the force constant of the harmonic spring, $\|\cdot\|$ denotes the Euclidian norm of a vector in $\mathcal{R}^2$, the functions $V_i : \mathcal{R} \to \mathcal{R}$, $i = 1, 2$, are assumed to be smooth but arbitrary otherwise, and $\boldsymbol{q}_i^0$, $i = 1, 2$, are two fixed reference vectors.

Let us introduce the following abbreviations:

$$\begin{aligned} \hat{r}_1 &:= \|\boldsymbol{q}_1 - \boldsymbol{q}_1^0\| \,, \\ \hat{r}_2 &:= \|\boldsymbol{q}_2 - \boldsymbol{q}_2^0\| \,, \\ r_{12} &:= \|\boldsymbol{q}_1 - \boldsymbol{q}_2\| \,. \end{aligned}$$

Then the equations of motion are

$$\frac{d}{dt}\boldsymbol{q}_1 = \boldsymbol{p}_1 \,, \tag{9a}$$

$$\frac{d}{dt}\boldsymbol{q}_2 = \boldsymbol{p}_2 \,, \tag{9b}$$

$$\frac{d}{dt}\boldsymbol{p}_1 = -\frac{V_1'(\hat{r}_1)}{\hat{r}_1}(\boldsymbol{q}_1 - \boldsymbol{q}_1^0) - \frac{\kappa}{r_{12}}(r_{12} - 1)(\boldsymbol{q}_1 - \boldsymbol{q}_2) \,, \tag{9c}$$

$$\frac{d}{dt}\boldsymbol{p}_2 = -\frac{V_2'(\hat{r}_2)}{\hat{r}_2}(\boldsymbol{q}_2 - \boldsymbol{q}_2^0) - \frac{\kappa}{r_{12}}(r_{12} - 1)(\boldsymbol{q}_2 - \boldsymbol{q}_1) \,. \tag{9d}$$

The qualitative solution behavior becomes more apparent when going to local coordinates, i.e., we rewrite the equations of motion in terms of the center of mass

$$\boldsymbol{q}_c = \frac{\boldsymbol{q}_1 + \boldsymbol{q}_2}{2} \,,$$

the internal bond stretching $r = r_{12}$, and the angle of rotation ϕ determined by

$$\boldsymbol{\psi}(\phi) = \begin{pmatrix} \cos\phi \\ \sin\phi \end{pmatrix} = \frac{\boldsymbol{q}_1 - \boldsymbol{q}_2}{r_{12}} \,.$$

This coordinate transformation gives rise to a corresponding transformation of the momenta via the canonical lift transformation [10]. Thus the corresponding conjugate momenta are $\boldsymbol{p}_c \in \mathcal{R}^2$, defined by

$$\boldsymbol{p}_c = \boldsymbol{p}_1 + \boldsymbol{p}_2 \, ,$$

and $p_r, p_\phi \in \mathcal{R}$, defined by[1]

$$\frac{\boldsymbol{p}_1 - \boldsymbol{p}_2}{2} = \frac{p_r}{r}(\boldsymbol{q}_1 - \boldsymbol{q}_2) + \frac{p_\phi}{r^2} J(\boldsymbol{q}_1 - \boldsymbol{q}_2) \tag{10}$$

where

$$J = \begin{bmatrix} 0 & -1 \\ 1 & 0 \end{bmatrix} .$$

To transform the Hamiltonian H into these new coordinates, we use the identities

$$\begin{aligned} \frac{1}{2}\boldsymbol{p}_1^T \boldsymbol{p}_1 + \frac{1}{2}\boldsymbol{p}_2^T \boldsymbol{p}_2 &= \frac{1}{4}(\boldsymbol{p}_1 + \boldsymbol{p}_2)^T(\boldsymbol{p}_1 + \boldsymbol{p}_2) + \frac{1}{4}(\boldsymbol{p}_1 - \boldsymbol{p}_2)^T(\boldsymbol{p}_1 - \boldsymbol{p}_2) \, , \\ &= \frac{1}{4}\boldsymbol{p}_c^T \boldsymbol{p}_c + p_r^2 + r^{-2} p_\phi^2 \end{aligned}$$

and

$$\begin{aligned} \boldsymbol{q}_1 &= \boldsymbol{q}_c + \frac{r}{2}\boldsymbol{\psi}(\phi) \, , \\ \boldsymbol{q}_2 &= \boldsymbol{q}_c - \frac{r}{2}\boldsymbol{\psi}(\phi) \, . \end{aligned}$$

Thus we obtain

$$H = \frac{\boldsymbol{p}_c^T \boldsymbol{p}_c}{4} + p_r^2 + r^{-2} p_\phi^2 + \frac{\kappa}{2}(r-1)^2 + V_1(\boldsymbol{q}_c, r, \phi, \boldsymbol{q}_1^0) + V_2(\boldsymbol{q}_c, r, \phi, \boldsymbol{q}_2^0) \, . \tag{11}$$

The corresponding equations of motion are

$$\frac{d}{dt}\boldsymbol{q}_c = \frac{1}{2}\boldsymbol{p}_c \, , \tag{12a}$$

$$\frac{d}{dt}\boldsymbol{p}_c = -\frac{V_1'(\hat{r}_1)}{\hat{r}_1}(\boldsymbol{q}_1 - \boldsymbol{q}_1^0) - \frac{V_2'(\hat{r}_2)}{\hat{r}_2}(\boldsymbol{q}_2 - \boldsymbol{q}_2^0) \tag{12b}$$

for the center of mass,

$$\frac{d}{dt} r = 2 p_r \, , \tag{12c}$$

$$\begin{aligned} \frac{d}{dt} p_r = -\kappa(r-1) + 2 p_\phi^2 r^{-3} - \frac{V_1'(\hat{r}_1)}{2\hat{r}_1}\left(\frac{r}{2} + \boldsymbol{\psi}^T[\boldsymbol{q}_c - \boldsymbol{q}_1^0]\right) + \\ - \frac{V_2'(\hat{r}_2)}{2\hat{r}_2}\left(\frac{r}{2} - \boldsymbol{\psi}^T[\boldsymbol{q}_c - \boldsymbol{q}_2^0]\right) \end{aligned} \tag{12d}$$

[1] Explicit expressions for p_r and p_ϕ are obtained by premultiplying (10) by $(\boldsymbol{q}_1 - \boldsymbol{q}_2)^T$, $(\boldsymbol{q}_1 - \boldsymbol{q}_2)^T J$ respectively.

for the internal bond vibrations, and

$$\frac{d}{dt}\phi = 2r^{-2}p_\phi\,, \tag{12e}$$

$$\frac{d}{dt}p_\phi = r\frac{V_1'(\hat{r}_1)}{2\hat{r}_1}\boldsymbol{\psi}^T J(\boldsymbol{q}_c - \boldsymbol{q}_1^0) - r\frac{V_2'(\hat{r}_2)}{2\hat{r}_2}\boldsymbol{\psi}^T J(\boldsymbol{q}_c - \boldsymbol{q}_2^0) \tag{12f}$$

for the rotation of the molecule. Here we have used

$$\nabla_x \hat{r}_i = \frac{1}{2}\hat{r}_i^{-1}\nabla_x \hat{r}_i^2, \qquad x = r, \phi, \boldsymbol{q}_c\,,$$

$$\hat{r}_i^2 = (\boldsymbol{q}_c - \boldsymbol{q}_i^0)^T(\boldsymbol{q}_c - \boldsymbol{q}_i^0) + \frac{1}{4}r^2 - (-1)^i r\,\boldsymbol{\psi}^T(\boldsymbol{q}_c - \boldsymbol{q}_i^0), \qquad i = 1, 2\,.$$

In the context of our semi-implicit methods, we typically consider the special case $V_1 = V_2 = 0$ which leads to the simplified equations of motion

$$\frac{d}{dt}\boldsymbol{q}_c = \frac{1}{2}\boldsymbol{p}_c\,, \tag{13a}$$

$$\frac{d}{dt}\boldsymbol{p}_c = \boldsymbol{0}\,, \tag{13b}$$

$$\frac{d}{dt}r = 2p_r\,, \tag{13c}$$

$$\frac{d}{dt}p_r = -\kappa(r-1) + 2p_\phi^2 r^{-3}\,, \tag{13d}$$

$$\frac{d}{dt}\phi = 2r^{-2}p_\phi\,, \tag{13e}$$

$$\frac{d}{dt}p_\phi = 0\,. \tag{13f}$$

4 Numerical Approximation

The standard discretization for the equations (9) in molecular dynamics is the (explicit) Verlet method. Stability considerations imply that the Verlet method must be applied with a step-size restriction $k < \varepsilon := \sqrt{2/\kappa}$. Various methods have been suggested to avoid this step-size barrier. The most popular is to replace the stiff spring by a holonomic constraint, as in (4). For our first model problem, this leads to the equations

$$\begin{aligned}
\frac{d}{dt}\boldsymbol{q}_1 &= \boldsymbol{p}_1\,,\\
\frac{d}{dt}\boldsymbol{q}_2 &= \boldsymbol{p}_2\,,\\
\frac{d}{dt}\boldsymbol{p}_1 &= -\frac{V_1'(\hat{r}_1)}{\hat{r}_1}(\boldsymbol{q}_1 - \boldsymbol{q}_1^0) - \frac{\lambda}{r_{12}}(\boldsymbol{q}_1 - \boldsymbol{q}_2)\,,\\
\frac{d}{dt}\boldsymbol{p}_2 &= -\frac{V_2'(\hat{r}_2)}{\hat{r}_2}(\boldsymbol{q}_2 - \boldsymbol{q}_2^0) - \frac{\lambda}{r_{12}}(\boldsymbol{q}_2 - \boldsymbol{q}_1)\,,\\
0 &= r_{12} - 1\,.
\end{aligned}$$

In local coordinates, the constraint is $r \equiv 1$, so equations (12) simplify directly into

$$
\begin{aligned}
\frac{d}{dt}\boldsymbol{q}_c &= \frac{1}{2}\boldsymbol{p}_c \,, \\
\frac{d}{dt}\boldsymbol{p}_c &= -\frac{V_1'(\hat{r}_1)}{\hat{r}_1}(\boldsymbol{q}_1 - \boldsymbol{q}_1^0) - \frac{V_2'(\hat{r}_2)}{\hat{r}_2}(\boldsymbol{q}_2 - \boldsymbol{q}_2^0) \,, \\
\frac{d}{dt}\phi &= 2p_\phi \,, \\
\frac{d}{dt}p_\phi &= \frac{V_1'(\hat{r}_1)}{2\hat{r}_1}\boldsymbol{\psi}^T J(\boldsymbol{q}_c - \boldsymbol{q}_1^0) - \frac{V_2'(\hat{r}_2)}{2\hat{r}_2}\boldsymbol{\psi}^T J(\boldsymbol{q}_c - \boldsymbol{q}_2^0) \,.
\end{aligned}
$$

The constrained equations of motion in cartesian coordinates can be solved by the SHAKE or (the essentially equivalent) RATTLE method (see [8]) which requires the solution of a non-linear system of equations in the Lagrange multiplier function λ. The equivalent formulation in local coordinates can still be integrated by using the explicit Verlet method.

The main disadvantage of this approach arises when the limit constrained system is different from (4), as mentioned in the introduction and demonstrated in §5 for our second model problem.

Another way to overcome the step-size restriction $k < \varepsilon$ is to use multiple-time-stepping methods [4] or implicit methods [17,18,12,3]. In this paper, we examine the latter possibility. But for large molecular systems, fully implicit methods are very expensive. For that reason, we focus on the general class of semi-implicit methods depicted in Fig. 1 [12]. In this scheme, Step 3 of the nth time step can be combined with Step 1 of the $(n+1)$st time step. This then is a staggered two-step splitting method. We refer to [12] for further justification.

Note that, in local coordinates, Step 2 is equivalent to integrating the equations (13). Thus, Step 2 can either be performed in local or in cartesian coordinates. We consider two different implicit methods for this purpose, namely, the midpoint method and the energy conserving method (6) which, in this example, coincides with the method (7) (because the V term appearing in (6) and (7) for $\mathbf{q} = \mathbf{q}_1 - \mathbf{q}_2$ is quadratic here). These methods are applied to the formulation in cartesian and in local coordinates and the properties of the resulting propagation maps are discussed next.

Semi-Implicit Integrator

Step 1.

$$\bar{\boldsymbol{p}}_{1,n} = \boldsymbol{p}_{1,n} - k\frac{V_1'(\hat{r}_{1,n})}{2\hat{r}_{1,n}}(\boldsymbol{q}_{1,n} - \boldsymbol{q}_1^0)\,,$$
$$\bar{\boldsymbol{p}}_{2,n} = \boldsymbol{p}_{2,n} - k\frac{V_2'(\hat{r}_{2,n})}{2\hat{r}_{2,n}}(\boldsymbol{q}_{2,n} - \boldsymbol{q}_2^0)\,.$$

Step 2.

Apply one step of size k to approximately integrate the fast system

$$\frac{d}{dt}\boldsymbol{q}_1 = \boldsymbol{p}_1\,,$$
$$\frac{d}{dt}\boldsymbol{q}_2 = \boldsymbol{p}_2\,,$$
$$\frac{d}{dt}\boldsymbol{p}_1 = -\frac{\kappa}{r_{12}}(r_{12}-1)(\boldsymbol{q}_1 - \boldsymbol{q}_2)\,,$$
$$\frac{d}{dt}\boldsymbol{p}_2 = -\frac{\kappa}{r_{12}}(r_{12}-1)(\boldsymbol{q}_2 - \boldsymbol{q}_1)$$

using an implicit method with initial conditions $(\boldsymbol{q}_{1,n}, \boldsymbol{q}_{2,n}, \bar{\boldsymbol{p}}_{1,n}, \bar{\boldsymbol{p}}_{2,n})$. Denote the result by $(\boldsymbol{q}_{1,n+1}, \boldsymbol{q}_{2,n+1}, \tilde{\boldsymbol{p}}_{1,n+1}, \tilde{\boldsymbol{p}}_{2,n+1})$.

Step 3.

$$\boldsymbol{p}_{1,n+1} = \tilde{\boldsymbol{p}}_{1,n+1} - k\frac{V_1'(\hat{r}_{1,n+1})}{2\hat{r}_{1,n+1}}(\boldsymbol{q}_{1,n+1} - \boldsymbol{q}_1^0)\,,$$
$$\boldsymbol{p}_{2,n+1} = \tilde{\boldsymbol{p}}_{2,n+1} - k\frac{V_2'(\hat{r}_{2,n+1})}{2\hat{r}_{2,n+1}}(\boldsymbol{q}_{2,n+1} - \boldsymbol{q}_2^0)\,.$$

Fig. 1. A semi-implicit integrator: the implicit scheme is applied only to the fast system.

4.1 Local Coordinates

The midpoint discretization of (13) yields

$$\boldsymbol{q}_{c,n+1} = \boldsymbol{q}_{c,n} + k\boldsymbol{p}_{c,n}/2\,, \tag{14a}$$
$$\boldsymbol{p}_{c,n+1} = \boldsymbol{p}_{c,n}\,, \tag{14b}$$
$$r_{n+1} = r_n + 2k\,p_{r,n+1/2}\,, \tag{14c}$$
$$p_{r,n+1} = p_{r,n} - k\kappa(r_{n+1/2} - 1) + kp_{\phi,n+1/2}^2 r_{n+1/2}^{-3}\,, \tag{14d}$$
$$\phi_{n+1} = \phi_n + 2kr_{n+1/2}^{-2}p_{\phi,n}\,, \tag{14e}$$
$$p_{\phi,n+1} = p_{\phi,n}\,. \tag{14f}$$

As remarked earlier, we are interested in the behavior of this approximation for step-sizes k much larger than the period of the fast bond vibrations, which is of order ε. But then, the Hamiltonian (11) is almost quadratic. The midpoint approximation almost reproduces this Hamiltonian [3,2], which provides an energy norm estimate for the approximate solution. A simple perturbation argument yields from this that the midpoint method applied to the local coordinates formulation of the fast system (13) is unconditionally stable and that, in the limit $\kappa \to \infty$, it conserves energy exactly. Note that, in local coordinates, the midpoint method and its energy conserving variants are basically equivalent.

4.2 Cartesian Coordinates

The interesting question is now what happens if the midpoint method is applied to the cartesian formulation (9) with $V_1 = V_2 = 0$. The equations are

$$\boldsymbol{q}_{1,n+1} = \boldsymbol{q}_{1,n} + k\boldsymbol{p}_{1,n+1/2}\,, \tag{15a}$$

$$\boldsymbol{q}_{2,n+1} = \boldsymbol{q}_{2,n} + k\boldsymbol{p}_{2,n+1/2}\,, \tag{15b}$$

$$\boldsymbol{p}_{1,n+1} = \boldsymbol{p}_{1,n} - \frac{k\,\kappa}{\tilde{r}_{n+1/2}}(\tilde{r}_{n+1/2} - 1)(\boldsymbol{q}_{1,n+1/2} - \boldsymbol{q}_{2,n+1/2})\,, \tag{15c}$$

$$\boldsymbol{p}_{2,n+1} = \boldsymbol{p}_{2,n} - \frac{k\,\kappa}{\tilde{r}_{n+1/2}}(\tilde{r}_{n+1/2} - 1)(\boldsymbol{q}_{2,n+1/2} - \boldsymbol{q}_{1,n+1/2})\,, \tag{15d}$$

where $\tilde{r}_{n+1/2} := \|\boldsymbol{q}_{1,n+1/2} - \boldsymbol{q}_{2,n+1/2}\|$. This discretization becomes unstable if the step-size k becomes too large [6]. As discussed in [3] by means of a simple linear time-varying problem, stability requires that

$$\alpha := k^2\sqrt{\kappa}$$

be sufficiently small. Since the midpoint discretization of the equations of motion in local coordinates (14) is stable for any step size k, the present instability might come as a surprise. The explanation can be given as follows [6,3]: In cartesian coordinates the fast vibrations and the slow translational and rotational degrees of freedom are not decoupled as they are in the local coordinate formulation. For large step sizes as compared to ε, this coupling leads to the destabilization of the midpoint method.

To obtain the unconditional stability of the midpoint method in local coordinates, one would have to consider the decoupling transformation from cartesian to local coordinates for the discrete variables $\boldsymbol{q}_{1,n}$ etc. But this transformation, which for the continuous variables is not constant, necessarily is in error which depends on k, not ε. The stability properties of the discrete dynamical systems obtained by the midpoint discretization in the different sets of coordinates may therefore be significantly different when $k \gg \varepsilon$ [3].

This instability is avoided when applying the method (6), as proved in [6]. The latter method yields here

$$\boldsymbol{q}_{1,n+1} = \boldsymbol{q}_{1,n} + k\boldsymbol{p}_{1,n+1/2}\,, \tag{16a}$$
$$\boldsymbol{q}_{2,n+1} = \boldsymbol{q}_{2,n} + k\boldsymbol{p}_{2,n+1/2}\,, \tag{16b}$$
$$\boldsymbol{p}_{1,n+1} = \boldsymbol{p}_{1,n} - \frac{k\,\kappa}{r_{n+1/2}}(r_{n+1/2}-1)(\boldsymbol{q}_{1,n+1/2} - \boldsymbol{q}_{2,n+1/2})\,, \tag{16c}$$
$$\boldsymbol{p}_{2,n+1} = \boldsymbol{p}_{2,n} - \frac{k\,\kappa}{r_{n+1/2}}(r_{n+1/2}-1)(\boldsymbol{q}_{2,n+1/2} - \boldsymbol{q}_{1,n+1/2})\,, \tag{16d}$$

where

$$r_{n+1/2} := \frac{1}{2}(\|\boldsymbol{q}_{1,n+1} - \boldsymbol{q}_{2,n+1}\| + \|\boldsymbol{q}_{1,n} - \boldsymbol{q}_{2,n}\|)\,.$$

The gain in stability can now be interpreted as resulting from the direct midpoint discretization of the rapidly vibrating, local variable r, thus avoiding the potentially damaging discrete decoupling transformation.

k	$\sqrt{\kappa}$	α	ΔH
0.05	0.5e+4	12.5	0.71e-3
0.05	1.0e+4	25	0.83e-3
0.1	0.5e+4	50	0.39e-2
0.1	1.0e+4	100	0.33e-2
0.2	0.5e+4	200	0.11e-1
0.2	1.0e+4	400	0.12e-1

Table 1. Maximum error in the energy using the semi-implicit method with the energy conserving method (6) for the strong forces.

Numerical Experiment. We now present numerical results for non-zero V_1 and V_2. In particular, we take

$$V_1(\hat{r}_1) := \frac{1}{2}(\hat{r}_1 - 1)^2\,, \qquad \hat{r}_1 = \|\boldsymbol{q}_1 - [1.5,0]^T\|\,,$$
$$V_2(\hat{r}_2) := \frac{1}{2}(\hat{r}_2 - 1)^2\,, \qquad \hat{r}_2 = \|\boldsymbol{q}_2 + [1.5,0]^T\|\,.$$

As initial conditions we chose $\boldsymbol{q}_1 = [0.6,0]^T$, $\boldsymbol{q}_2 = [-0.4,0]^T$, $\boldsymbol{p}_1 = [1,1]^T$, and $\boldsymbol{p}_2 = [-1,-1]^T$.

We apply the semi-implicit algorithm to handle the weak potentials V_i, and the energy conserving method (16) for the stiff forces. The maximal error in the total energy, i.e.

$$\Delta H = \max_{t\in[0,500]} |H(t) - H(0)|\,,$$

can be found in Table 1 for various values of k, κ, and α. Even for large κ and α, the value of ΔH depends quadratically on the step size k.

A difficulty with the energy conserving method (6), in general, is the solution of the corresponding nonlinear equations [6]. Here, however, using the initial iterate $(\mathbf{q}_n + k\mathbf{p}_n, \mathbf{p}_n)$ for $(\mathbf{q}_{n+1}, \mathbf{p}_{n+1})$, even for large values of α we did not observe any difficulties with the convergence of Newton's method.

On the other hand, our computations using (15) indicate that the midpoint method becomes unstable for $\alpha > 1$.

Next, we replace the stiff spring potential $\kappa(r-1)^2/2$ by the Morse potential

$$\kappa(1 - e^{-(r-1)/2})^2,$$

where $r = \|\mathbf{q}_1 - \mathbf{q}_2\|$ as before. The methods (6) and (7) are no longer the same, and (7) does not conserve the energy exactly, even in the absence of the slow potentials. However, these two methods remain close. Repeating the experiments of Table 1 produces qualitatively similar results for the Morse potential. The computations with either (6) or (7) are stable. The energy error, as well as the error in fast energy ΔE_f defined in the next section, appear to converge quadratically in k and be independent of κ, when $k \gg \sqrt{\kappa}$. Similar numerical experiments with the midpoint method run into difficulties.

5 Modified Model Problem

For molecules with more than two atoms the frequencies of the bond stretching modes and/or bond angle bending modes are, in general, no longer constant along the slowly varying solution components. This implies additional complications not present for our simple diatomic model problem. In particular, in the limit $\kappa \to \infty$, replacing the stiff spring terms by holonomic constraints as in (4) leads to a qualitatively wrong dynamics which has to be corrected by introducing an additional force term [14,5]. The size of this additional force term depends on the limiting initial conditions.

Unfortunately, discretization methods with large step sizes applied to such problems tend to miss this additional force term [3]. Furthermore, even if the implicit midpoint method is applied to a formulation in local coordinates, similar problems occur [3]. Since the midpoint scheme and its variants (6) and (7) are basically identical in local coordinates, the same problem can be expected for the energy conserving method (6). To demonstrate this, let us consider the following modified model problem [13]:

In the model problem described earlier, replace the stiff spring potential $V_s = \kappa(r-1)^2/2$ by

$$V_s = \frac{\kappa}{2}(\|A(\boldsymbol{q}_1 - \boldsymbol{q}_2)\| - 1)^2$$

with

$$A = \begin{bmatrix} \sqrt{2} & 0 \\ 0 & 1 \end{bmatrix} .$$

This implies that the two "atoms" oscillate about a mutual distance $d = r(\phi)$ that depends on the angle ϕ and is given by an ellipse. Let

$$d = \| A(\boldsymbol{q}_1 - \boldsymbol{q}_2) \| .$$

The equations of motion are an obvious modification of (9) where $\boldsymbol{q}_1 - \boldsymbol{q}_2$ is replaced by $A^2(\boldsymbol{q}_1 - \boldsymbol{q}_2)$ and r_{12} is replaced by d. The total energy H of the system decomposes into the vibrational energy E_f of the stiff "spring" and the energy in the slowly varying degrees of freedom. The time-evolution of $E_f(t)$ is crucial for the limiting behavior of (3a)-(3b) as $\kappa \to \infty$. Let us derive an explicit expression for the energy E_f. The conjugate momentum corresponding to d is

$$p_d := \frac{d}{2\|A^2(\boldsymbol{q}_1 - \boldsymbol{q}_2)\|^2} (\boldsymbol{p}_1 - \boldsymbol{p}_2)^T A^2 (\boldsymbol{q}_1 - \boldsymbol{q}_2) .$$

The vibrational energy E_f is now given by

$$E_f = \omega^2(\boldsymbol{q}_1 - \boldsymbol{q}_2)\, p_d^2 + \frac{\kappa}{2}(d-1)^2$$

with

$$\omega(\boldsymbol{q}_1 - \boldsymbol{q}_2) := \frac{\|A^2(\boldsymbol{q}_1 - \boldsymbol{q}_2)\|}{d} .$$

Note that E_f corresponds to a high-frequency harmonic oscillator with a slowly varying frequency ω. This implies that the vibrational energy E_f is not constant along solution curves. Instead, we have the adiabatic invariant $J_f := E_f/\omega$ which is preserved up to terms of order $\varepsilon = \sqrt{2/\kappa}$ over a time-interval of order at least one. In the limit $\kappa \to \infty$, the slow motion is no longer given by simply enforcing $d - 1 = 0$ as a holonomic constraint. Instead, a force term corresponding to the additional potential energy

$$V_c = J_f \omega(\boldsymbol{q}_1 - \boldsymbol{q}_2)$$

has to be added in (4).

Numerical Experiment. We consider the case $V_1 = V_2 = 0$ and apply the energy conserving method (6). (Note that the methods (6) and (7) are still equivalent, with the corresponding notation $r \leftarrow d$, $V(d) \leftarrow \frac{\kappa}{2}(d-1)^2$, and $\mathbf{q} \leftarrow A^2(\mathbf{q}_1 - \mathbf{q}_2)$.) As initial values we took $\boldsymbol{q}_1 = [0, 0.5]^T$, $\boldsymbol{q}_2 = [0, -0.5]^T$, $\boldsymbol{p}_1 = [1, 1]^T$, and $\boldsymbol{p}_2 = [-1, -1]^T$. The maximal variation in the vibrational energy, i.e.

$$\Delta E_f = \max_{t \in [0,10]} |E_f(t) - E_f(0)| ,$$

k	κ	$k\sqrt{\kappa}$	ΔE_f	ΔJ_f
0.1e-2	1.0e+4	0.1	0.42	0.57e-1
0.1e-1	1.0e+4	1	0.24	0.14
0.5e-1	1.0e+4	5	0.59e-1	0.29
0.1e-3	1.0e+6	0.1	0.41	0.57e-2
0.1e-2	1.0e+6	1	0.23	0.13
0.1e-1	1.0e+6	10	0.62e-2	0.29
0.2e-1	1.0e+6	20	0.60e-2	0.29

Table 2. Maximum variation in the vibrational energy E_f and the adiabatic invariant J_f using the energy conserving method (6).

and in the adiabatic invariant, i.e.

$$\Delta J_f = \max_{t\in[0,10]} |J_f(t) - J_f(0)|\,,$$

can be found in Table 2. Note that, for $k\sqrt{\kappa} \gg 1$, the method enforces $\Delta E_f \approx 0$ (and thus $V_c = 0$) instead of (approximately) conserving the adiabatic invariant J_f. This implies that the energy conserving method introduces an error of order one in the slow solution quantities, unless $k\sqrt{\kappa}$ is bounded by a constant of magnitude order 1.[2] This is a step-size restriction which is comparable to the explicit Verlet method stability restriction, up to a moderate factor. In fact, the energy conserving method leads to the same wrong solution behavior as a naive enforcement of the holonomic constraint $d = 1$ by a method such as SHAKE. Note that, due to the energy conserving nature of the method, no indication of this wrong solution behavior will be given unless the adiabatic invariant is explicitly computed. Again we did not observe any difficulties in solving the nonlinear equations by Newton's method, so no alarm bells of any kind rang while computing a wrong solution.

References

1. M.P. Allen and T.J. Tildesley. *Computer Simulations of Liquids*. Clarendon Press, Oxford, 1987.
2. U. Ascher. Stabilization of invariants of discretized differential systems. *Numerical Algorithms*, 14:1–23, 1997.
3. U. Ascher and S. Reich. The midpoint scheme and variants for Hamiltonian systems: advantages and pitfalls. 1996. submitted.
4. J.J. Biesiadecki and R.D. Skeel. Danger of multiple-time-step methods. *J. Comput. Phys.*, 109:318–328, 1993.

[2] To be more precise, this error occurs in the limit $\kappa \to \infty$ with $E_f = \mathcal{O}(1)$ and step-size k such that $k\sqrt{\kappa} = const. \gg 1$. This error does not occur if $E_f = 0$ for the analytic problem, i.e., in case there is no vibrational energy in the stiff "spring" which implies $V_c = 0$.

5. F.A. Bornemann and Ch. Schutte. Homogenization of Hamiltonian systems with a strong constraining potential. *Physica D*, 102:57–77, 1997.
6. O. Gonzalez and J. Simo. On the stability of symplectic and energy-momentum algorithms for nonlinear Hamiltonian systems with symmetry. *Comp. Meth. in Appl. Mech. Eng.*, 134:197–222, 1996.
7. E. Hairer and G. Wanner. *Solving Ordinary Differential Equations II: Stiff and Differential-Algebraic Problems.* Springer-Verlag, 1991.
8. B. Leimkuhler and R. Skeel. Symplectic integrators in constrained Hamiltonian systems. *J. Comp. Phys.*, 112:117–125, 1994.
9. M. Mandziuk and T. Schlick. Resonance in the dynamices of chemical systems simulated by the implicit midpoint scheme. *Chem. Phys. Lett.*, 237:525–535, 1995.
10. J.E. Marsden and T.S. Ratiu. *Introduction to Mechanics and Symmetry.* Springer-Verlag, New York, 1994.
11. L.R. Petzold, L.O. Jay, and J. Yen. Numerical solution of highly oscillatory ordinary differential equations. *Acta Numerica*, pages 437–484, 1997.
12. S. Reich. Enhanced energy conserving methods. *BIT*, 36:122–134, 1996.
13. S. Reich. Preservation of adiabatic invariants under symplectic discretization. *App. Numer. Math.*, to appear, 1998.
14. H. Rubin and P. Ungar. Motion under a strong constraining force. *Comm. Pure Appl. Math.*, 10:65–87, 1957.
15. J.C. Simo and O. Gonzales. Assessment of energy-monentum and symplectic schemes for stiff dynamical systems. *The American Society of Mechanical Engineering*, 93-WA/PVP-4, 1993.
16. T. Schlick, M. Mandziuk, R.D. Skeel, and K. Srinivas. Nonlinear resonance artifacts in molecular dynamics simulations. *J. Comput. Phys.*, 139:1–29, 1998.
17. R.D. Skeel, G. Zhang, and T. Schlick. A family of symplectic integrators: stability, accuracy, and molecular dynamics applications. *SIAM J. Sci. Comput.*, 18:203–222, 1997.
18. R.D. Skeel and M. Zhang. Cheap implicit symplectic integrators. *Appl. Numer. Math.*, 25:297–302, 1997.
19. L. Verlet. Computer experiments on classical fluids. *Phys. Rev.*, 159:98–103, 1967.