Smoothed Dynamics of Highly Oscillatory Hamiltonian Systems*

Sebastian Reich
Konrad-Zuse Zentrum Berlin
Heilbronner Str. 10
D-10711 Berlin

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Abstract

We consider the numerical treatment of Hamiltonian systems that contain a potential which grows large when the system deviates from the equilibrium value of the potential. Such systems arise, e.g., in molecular dynamics simulations and the spatial discretization of Hamiltonian partial differential equations. Since the presence of highly oscillatory terms in the solutions forces any explicit integrator to use very small step-size, the numerical integration of such systems provides a challenging task. It has been suggested before to replace the strong potential by a holonomic constraint that forces the solutions to stay at the equilibrium value of the potential. This approach has, e.g., been successfully applied to the bond stretching in molecular dynamics simulations. In other cases, such as the bond-angle bending, this methods fails due to the introduced rigidity. Here we give a careful analysis of the analytical problem by means of a smoothing operator. This will lead us to the notion of the smoothed dynamics of a highly oscillatory Hamiltonian system. Based on our analysis, we suggest a new constrained formulation that maintains the flexibility of the system while at the same time suppressing the high-frequency components in the solutions and thus allowing for larger time steps. The new constrained formulation is Hamiltonian and can be discretized by the well-known SHAKE method.

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1 Introduction

We are concerned in this paper with the numerical solution of singularly perturbed Hamiltonian systems of the form

$$\begin{aligned} \frac{d}{dt}q &= M^{-1}p \\ \frac{d}{dt}p &= -\nabla V(q) - \frac{1}{\epsilon^2}G(q)^T K g(q) \end{aligned} \tag{1}$$

with Hamiltonian function

$$H(q,p) = \frac{p^T M^{-1} p}{2} + V(q) + \frac{1}{\epsilon^2}\frac{g(q)^T K g(q)}{2} \tag{2}$$

where ϵ a small parameter, $q, p \in R^n$, and $G(q) = g_q(q)$. Here M is the positive definite mass matrix of the system, $V : R^n \to R$ the potential energy function, and g is the collection of functions $g_i : R^n \to R$, $i = 1, \ldots, m$, with corresponding (scaled) force constant $K_{i,i}$, i.e.

$$\frac{g(q)^T K g(q)}{2} = \frac{1}{2}\sum_i K_{i,i}\, g_i(q)^2$$

and K the m-dimensional diagonal matrix with entries $K_{i,i}$.

Note that the parameter ϵ has no immediate physical meaning and is not uniquely determined by the physical problem. It stands for the fact that the potential $g(q)^T K g(q)/(2\epsilon^2)$ grows large away from its equilibrium value $g(q) = 0$ compared to $V(q)$ and it allows one to treat the mathematical consequences of this fact in a relatively elegant way.

Throughout the paper we will use the following convention: Assume that the Hamiltonian (2) has been scaled such that

$$||V_{qq}(q)|| \leq 1 \tag{3}$$

Then ϵ is chosen such that

$$||K^{-1}|| \leq 1 \tag{4}$$

where we assume that the thus defined ϵ satisfies $\epsilon \ll 1$.

Let us demonstrate this for the quadratic Hamiltonian

$$H(q,p) = \frac{p^T M^{-1} p}{2} + \frac{q^T W q}{2} + \frac{q^T G^T U G q}{2}$$

where W is a positive definite $n \times n$ matrix, G is a $n \times m$ matrix, $m < n$, and U is a diagonal $m \times m$ matrix. Here we would first premultiply the whole Hamiltonian by $1/||W||$ and define ϵ by

$$\epsilon := \sqrt{||W||\,||U^{-1}||}$$

Thus ϵ is proportional to the ratio of the largest eigenvalue of W to the smallest eigenvalue of U. Provided that $\epsilon \ll 1$, we would then define the function g in (2) by

$$g(q) \quad := \quad Gq$$

and the matrix K by

$$K \quad := \quad ||U^{-1}||\, U$$

Hamiltonian systems of type (1) arise typically in the context of molecular dynamics simulations [10] (which provides the main motivation of this paper) and in the spatial discretization of Hamiltonian (hyperbolic) PDEs [9] like, for example, the Sine-Gordon equation by spectral or related methods. In the context of molecular dynamics, the potential $g(q)^T K g(q)/(2\epsilon^2)$ stands for covalent bond stretching and/or bond-angle bending; i.e., $g_i(q) = r - r_0$ and $\epsilon^2 \approx 0.01$ in case of bond stretching and $g_i(q) = \phi - \phi_0$ and $\epsilon^2 \approx 0.1$ in case of bond-angle bending. In the context of hyperbolic PDEs, the same expression is related to the high frequency modes in the Fourier spectrum of the solutions.

Differential equations of the form (1) fall into the class of singularly perturbed systems of type [4]

$$\frac{d}{dt} z \quad = \quad \frac{1}{\epsilon} f(z, \epsilon) \tag{5}$$

(see Section 2 for more details). Solutions of (5) satisfy, in general,

$$|z(t)| \quad = \quad O(1)$$

and

$$|\frac{d}{dt} z(t)| \quad = \quad O(\epsilon^{-1})$$

i.e., they are bounded but vary rapidly in t. Thus the step-size of a numerical integrator has, in general, to be of order $O(\epsilon)$. This implies a significant amount of computational work for the numerical integration over time intervals of order $O(1)$. For example, the lengths of a molecular dynamics simulation with an explicit method like Verlet [23] is for that reason restricted to a few tens of picoseconds up to a few nanoseconds, depending on the size of the problem [10]. This means that the time scale of the process that can be simulated is limited. To simulate processes over longer periods of time, new integration methods are essential.

Most of the theory has been developed for singularly perturbed problems that satisfy

1. rank $f_z(z,0) = \text{const.}$

for all z. This implies that the set $\mathcal{M}_0$ defined by

$$\mathcal{M}_0 \ := \ \{z : f(z,0) = 0\}$$

is a smooth manifold. The more stringent requirement is however that

2. $\mathcal{M}_0$ is an exponentially stable manifold of the differential equation

$$\frac{d}{dt} z \ = \ f(z,0)$$

Under the Assumptions 2, one can show that there exists a family $\mathcal{M}_\epsilon$ of smooth manifolds with $\mathcal{M}_{\epsilon=0} = \mathcal{M}_0$ such that $\mathcal{M}_\epsilon$ is an exponentially stable invariant manifold of (5) [4]. Furthermore, the solutions on $\mathcal{M}_\epsilon$ reflect the long-time behavior of the general solutions of (5) with initial values in a δ-neighborhood of $\mathcal{M}_\epsilon$ up to terms of order $O(\delta\,\epsilon)$. Since the solutions on $\mathcal{M}_\epsilon$ satisfy now $dz/dt = O(1)$, time-steps of order $O(1)$ can be used in a numerical integrator provided that the equations are discretized by a proper (implicit) method [11].

However, Assumption 2 is not satisfied for singularly perturbed Hamiltonian systems. In particular, solutions of (1) oscillate highly about the manifold $\mathcal{M}_0$. Thus, as we will show in Section 2, the manifold $\mathcal{M}_0$ does not even satisfy the weaker assumption of normal-hyperbolicity [4],[6]. This leaves us with the task of finding a different approach to the long-time integration of (1). In this paper, we attempt to do so by introducing the notion of the *smoothed dynamics* of highly oscillatory Hamiltonian systems. By this we mean the following:

Because of (3), the shortest period in the motion of (1) due to the potential $V(q)$ is of order $O(1)$. In contrast to this, the potential $g(q)^T K g(q)/(2\epsilon^2)$ contributes high-frequency terms with period of order $O(\epsilon)$. To separate these high frequency components from the slowly varying parts, we introduce the smoothing operator

$$\langle w \rangle_\alpha (t) \ := \ \frac{1}{\alpha} \int_{-\infty}^{+\infty} \rho(\frac{t-t'}{\alpha})\, w(t')\, dt' \tag{6}$$

with $0 < \alpha \ll 1$ and $w : \boldsymbol{R} \to \boldsymbol{R}$. Here $\rho : \boldsymbol{R} \to \boldsymbol{R}$ is an appropriate weight function such that for any (bounded) continuous function w there is a smooth (C^∞) function $\bar{w}$ with

$$\langle w \rangle_\alpha(t) - \bar{w}(t) \ = \ O(\alpha^s) \tag{7}$$

and for any smooth (C^∞) function w we have

$$\langle w \rangle_\alpha(t) - w(t) \ = \ O(\alpha^s) \tag{8}$$

where s is a fixed integer with $s \gg 1$. One could, for example, chose for ρ the Meyer scaling function [3]. Note that, in the frequency domain, the smoothing operator (6) corresponds to a low pass filter with cut-off frequency $\omega_c = O(1/\alpha)$.

The idea is now to replace the rapidly varying solutions $q(t)$ of (1) by $\langle q\rangle_\alpha(t)$ with

$$\alpha \;=\; \sqrt{\epsilon}$$

and then to seek numerical approximations to the smooth $\langle q\rangle_{\sqrt{\epsilon}}$ rather then to the rapidly varying $q(t)$. We call the functions $\langle q\rangle_{\sqrt{\epsilon}}(t)$, corresponding to solutions $q(t)$ of (1), the *smoothed dynamics* of (1).

We will discuss the properties of (6) in more detail in Section 2. There we will also show how to reformulate (1) as a singularly perturbed problem (5). In Section 3, we will then derive a constrained Hamiltonian system that approximates the smoothed dynamics of (1).

The approximation of (1) by a constrained Hamiltonian systems has been considered before (see, for example, [16],[22]). In a naive approach, one would introduce the new variable

$$\lambda \;:=\; \frac{1}{\epsilon^2}\,Kg(q)$$

and rewrite (1) as

$$\begin{aligned} \frac{d}{dt}q &= M^{-1}p \\ \frac{d}{dt}p &= -\nabla V(q) - G(q)^T\lambda \\ \epsilon^2 K^{-1}\lambda &= g(q) \end{aligned} \tag{9}$$

In the limit $\epsilon \to 0$, we obtain the constrained system

$$\begin{aligned} \frac{d}{dt}q &= M^{-1}p \\ \frac{d}{dt}p &= -\nabla V(q) - G(q)^T\lambda \\ 0 &= g(q) \end{aligned} \tag{10}$$

which is Hamiltonian on the constrained manifold

$$\mathcal{M}_0 \;=\; \{(q,p) : g(q) = 0,\, G(q)M^{-1}p = 0\,\} \tag{11}$$

provided that the matrix

$$G(q)M^{-1}G(q)^T \tag{12}$$

is invertible [9].

The solutions on $\mathcal{M}_0$ are now smooth. However, for initial values in a δ-neighborhood of $\mathcal{M}_0$, the approximation (5) introduces an error of order $O(\delta)$ over bounded time intervals (see Section 3). While this error is, for example, not significant for the covalent bond stretching in molecular dynamics simulations where $\delta \approx 0.01$, the same formulation (10) yields qualitatively wrong results when applied to the bond-angle bending or the harmonic dihedral bending where $\delta \approx 0.1$. The constrained formulation derived in this paper approximates the smoothed dynamics of (1) up to terms of order $O(\delta\, \epsilon^2)$ and provides therefore a qualitative improvement over (10). Finally, in Section 4, we discuss various numerical aspects of our new method and demonstrate its properties by means of two simple numerical examples.

Another approach to the long-time integration of highly oscillatory Hamiltonian system has been taken by Simo and his collaborators [20]. They advocate the direct discretization of (1) by an implicit energy-momentum method and the usage of a large step-size. However, we are not aware of rigorous stability and convergence results for these methods when applied to the system (1) with a step-size $\Delta t \gg \epsilon$.

2 Mathematical Background

In the first part of this section we show how to reformulate (1) as a singularly perturbed problem (5). To do so, we introduce local coordinates (q_1, q_2) by

$$\begin{aligned} q_1 &= g(q) \\ q_2 &= b(q) \end{aligned}$$

where $b(q)$ is a vector valued function such that $B(q)M^{-1}G(q)^T = 0$, $B(q) = b_q(q)$, and the composed matrix $[G(q)^T\, B(q)^T]$ is invertible. The existence of such a coordinate system follows, at least locally, from the Frobenius Theorem [1]. The corresponding conjugate momenta are given by

$$[G(q)^T\, B(q)^T] \begin{bmatrix} p_1 \\ p_2 \end{bmatrix} = p$$

which results in the Hamiltonian

$$H(q,p) = \frac{p_1^T G M^{-1} G^T p_1}{2} + \frac{p_2^T B M^{-1} B^T p_2}{2} + V + \frac{1}{\epsilon^2}\frac{q_1^T K q_1}{2} \qquad (13)$$

The equations of motion are now given by

$$\begin{aligned} \frac{d}{dt} q_1 &= G M^{-1} G^T p_1 \\ \frac{d}{dt} p_1 &= -\nabla_{q_1} V - \frac{1}{\epsilon^2} K q_1 - \nabla_{q_1} \frac{p_1^T G M^{-1} G^T p_1 + p_2^T B M^{-1} B^T p_2}{2} \end{aligned} \qquad (14)$$

and

$$\begin{aligned} \frac{d}{dt} q_2 &= BM^{-1}B^T\, p_2 \\ \frac{d}{dt} p_2 &= -\nabla_{q_2} V - \nabla_{q_2} \frac{p_1^T GM^{-1}G^T p_1 + p_2^T BM^{-1}B^T p_2}{2} \end{aligned} \tag{15}$$

where, for notational convenience, we suppressed the arguments in the mappings $V(q_1, q_2)$, $G(q_1, q_2)$, and $B(q_1, q_2)$.

Upon rescaling p_1 in (14) by ϵ, the equations (14) become

$$\begin{aligned} \frac{d}{dt} q_1 &= \frac{1}{\epsilon} GM^{-1}G^T\, p_1 \\ \frac{d}{dt} p_1 &= -\epsilon\, \nabla_{q_1} V - \frac{1}{\epsilon} K q_1 - \nabla_{q_1} \frac{p_1^T GM^{-1}G^T p_1 + \epsilon^2\, p_2^T BM^{-1}B^T p_2}{2\,\epsilon} \end{aligned} \tag{16}$$

which are now of the form (5). The corresponding manifold $\mathcal{M}_0$ is given by $q_1 = p_1 = 0$ or, in the original variables by (11). Linearization of (16) about the manifold $\mathcal{M}_0$ yields a linear system with eigenvalues on the imaginary axis. Thus the manifold $\mathcal{M}_0$ is not normally hyperbolic and the persistence of $\mathcal{M}_o$ for $\epsilon > 0$ cannot be concluded [6].

Let us now review a few results from statistical mechanics. Under the assumption that a given Hamiltonian system is ergodic, equipartition of energy [12] implies that

$$\langle p^i \frac{\partial H}{\partial p^i} \rangle = \delta \tag{17}$$

and

$$\langle q^i \frac{\partial H}{\partial q^i} \rangle = -\delta \tag{18}$$

where q^i and p^i, $i = 1, \ldots, n$, denote the ith component of the vector q, p respectively. Here H is the Hamiltonian of the system, δ corresponds to $k_B T$ in statistical mechanics; T the temperature and k_B the Boltzmann constant, and $\langle w \rangle$ denotes the time-average of a quantity $w(t)$; i.e.

$$\langle w \rangle := \lim_{T \to \infty} \frac{1}{2T} \int_{-T}^{T} w(t)\, dt \tag{19}$$

Throughout this paper, we will always assume that, for the given scaling of the Hamiltonian (2),

$$\epsilon^2 \leq \delta < 1 \tag{20}$$

For example, in molecular dynamics simulations $k_B T$ corresponds at room-temperature to $\delta \approx 0.01$ if the function g in (2) includes only the bond-stretching potentials and to $\delta \approx 0.1$ if g includes also the bond-bending potentials. Note that the different values of δ are due to the fact that we always scale the Hamiltonian (2) in such a way that the potential $V(q)$ satisfies (3).

As a consequency of (17), we obtain for the Hamiltonian (13) that

$$\langle p_1^T G(q) M^{-1} G(q)^T p_1 \rangle \quad = \quad m\,\delta\,, \tag{21}$$

and

$$\langle p_2^T B(q) M^{-1} B(q)^T p_2 \rangle \quad = \quad (n-m)\,\delta \tag{22}$$

where m is the dimension of the vector valued function g.

Now we want to derive a few important properties of the smoothing operator (6). We assume that $\rho : R \to R$ is a smooth function that goes to zero, as $|t| \to \infty$, faster than any inverse power of t, $\rho(0) = 1$, and

$$\int_{-\infty}^{\infty} \rho(t) dt \quad = \quad 1$$

The proper construction of a ρ, such that in addition (7) and (8) hold, falls into the subject of filter design and wavelet analysis [3].

The following four propositions will be crucial for the derivation of the smoothed dynamics of (1).

Proposition 1. *Let $w(t)$ be a differentiable function, then*

$$\langle \frac{d}{dt} w \rangle_\alpha \quad = \quad \frac{d}{dt} \langle w \rangle_\alpha$$

Proof. We have

$$\begin{aligned} \frac{d}{dt}\langle w \rangle_\alpha (t) \quad &= \quad \frac{d}{dt}\frac{1}{\alpha}\int_{-\infty}^{+\infty} \rho(\frac{t-t'}{\alpha})\, w(t')\, dt' \\ &= \quad \frac{1}{\alpha}\int_{-\infty}^{+\infty} -\frac{d}{dt'}\rho(\frac{t-t'}{\alpha})\, w(t')\, dt' \end{aligned}$$

Now, since $\rho(\pm\infty) = 0$, integration by parts yields

$$\begin{aligned} \frac{1}{\alpha}\int_{-\infty}^{+\infty} -\frac{d}{dt'}\rho(\frac{t-t'}{\alpha})\, w(t')\, dt' \quad &= \quad \frac{1}{\alpha}\int_{-\infty}^{+\infty} \rho(\frac{t-t'}{\alpha})\, \frac{d}{dt'} w(t')\, dt' \\ &= \quad \langle \frac{d}{dt} w \rangle_\alpha (t) \end{aligned}$$

Proposition 2. *Let $w(t)$ be an arbitrary (scalar valued) function such that*

$$(\langle w\rangle_\alpha)^k \quad = \quad \langle w^k\rangle_\alpha + O(\alpha^l)$$

for $k = 2, 3, \ldots$, then

$$\langle f(w)\rangle_\alpha \quad = \quad f(\langle w\rangle_\alpha) + O(\alpha^l)$$

where $f : R \to R$ is a smooth function and l a positive real integer.

Proof. Taylor expansion of f yields immediately the desired result.

Proposition 3. *Let $w(t)$ be an arbitrary (scalar valued) function. We write $w(t)$ as*

$$w(t) \quad = \quad w_a(t) + w_f(t)$$

where $w_a := \langle w\rangle_\alpha$. Then, up to terms of order $O(\alpha^s)$,

$$\langle w^2\rangle_\alpha\,(t) \quad = \quad w_a^2(t) + \langle w_f^2\rangle_\alpha(t)$$

Proof. Since, up to terms of order $O(\alpha^s)$, $\langle w_a w_f\rangle_\alpha = w_a\langle w_f\rangle_\alpha = 0$, $\langle w_a^2\rangle_\alpha = w_a^2$.

Proposition 4. *Let w_f be defined as in Proposition 3. Assume that the scaled function $\bar{w}_f(t) = w_f(\alpha^2 t)$ is smooth. Then*

$$\lim_{\alpha\to 0}\langle w_f^2\rangle_\alpha(t) \quad = \quad \langle \bar{w}_f^2\rangle$$

Proof. Let $\tau = t/\alpha^2$ and $\tau' = t'/\alpha^2$. Then

$$\begin{aligned}
\lim_{\alpha\to 0}\frac{1}{\alpha}\int_{-\infty}^{+\infty}\rho(\frac{t-t'}{\alpha})\, w_f^2(t')\,dt' &= \lim_{\alpha\to 0}\alpha\int_{-\infty}^{+\infty}\rho(\alpha[\tau-\tau'])\,\bar{w}_f^2(\tau')\,d\tau' \\
&= \lim_{\alpha\to 0}\frac{\alpha}{2}\int_{-\alpha/2}^{+\alpha/2}\rho(0)\,\bar{w}_f^2(\tau')\,d\tau' \\
&= \langle \bar{w}_f^2\rangle
\end{aligned}$$

Finally we want to apply Proposition 3 and 4 to the equations (14) and (15). In the limit $\alpha = \sqrt{\epsilon} \to 0$, we obviously have $q_{2,f} = p_{2,f} = 0$ while $q_{1,f}$ and $p_{1,f}$ satisfy the differential equation

$$\begin{aligned}
\frac{d}{dt}q_1 &= GM^{-1}G^T p_1 \\
\frac{d}{dt}p_1 &= -\frac{1}{\epsilon^2}Kq_1 - \nabla_{q_1}\frac{p_1^T GM^{-1}G^T p_1}{2}
\end{aligned}$$

Thus the scaled solutions $q_1(\epsilon t)$ and $p_1(\epsilon t)$ are smooth and Proposition 4, together with (18), implies

$$\begin{aligned}\lim_{\sqrt{\epsilon}\to 0} \langle q_{1,f}^T\, q_{1,f}\rangle_{\sqrt{\epsilon}} &= \langle q_{1,f}^T\, q_{1,f}\rangle \\ &= O(\delta\, \epsilon^2)\end{aligned}$$

A similar result can be derived for $\langle p_{1,f}^T\, p_{1,f}\rangle_{\sqrt{\epsilon}}$.

Let us turn now to the quantities $q_{1,a}$ and $p_{1,a}$. Since the energy per degree of freedom has to remain bounded as $\epsilon \to 0$, we have

$$q_{1,a} = O(\epsilon^2)$$

which also implies that $p_{1,a} = O(\epsilon^2)$, $dq_{1,a}/dt = O(\epsilon^2)$, and $dp_{1,a}/dt = O(\epsilon^2)$. Putting these results together, Proposition 3 yields, e.g., for the variable q_1 that

$$\langle q_1^T\, q_1\rangle_{\sqrt{\epsilon}} = O(\delta\, \epsilon^2) \tag{23}$$

3 Smoothed Dynamics – Analytical Results

In this section we show how the application of the smoothing operator (6) to (1) leads to constrained equations of motion that approximate the smoothed dynamics of (1). By such an approximation we mean a (constrained) Hamiltonian system with a Hamiltonian $H_e(Q, P)$ such that the corresponding solutions $(Q(t), P(t))$ satisfy

$$\langle q\rangle_{\sqrt{\epsilon}}(t) - Q(t) = O(\delta^k\, \epsilon^l)$$

and

$$\langle p\rangle_{\sqrt{\epsilon}}(t) - P(t) = O(\delta^k\, \epsilon^l)$$

over bounded intervals of time. Here $l > 0$ and $k \geq 0$ are appropriate integers, $(q(t), p(t))$ is a solution of (1), and $(Q(0), P(0))$ is chosen such that $\langle q\rangle_{\sqrt{\epsilon}}(0) - Q(0) = O(\delta^k\, \epsilon^l)$ and $\langle p\rangle_{\sqrt{\epsilon}}(0) - P(0) = O(\delta^k\, \epsilon^l)$. We will first derive an order $O(\epsilon^2)$ approximation and then improve this formulation to order $O(\delta\, \epsilon^2)$.

Remark. In [7], Kreiss introduced the concept of *slow solutions* for singularly perturbed systems (5). In our notation a slow solution of (1) is a solution $(q(t), p(t))$ that satisfies $(q(t), p(t)) \approx (\langle q\rangle_{\sqrt{\epsilon}}(t), \langle p\rangle_{\sqrt{\epsilon}}(t))$. We will see below that the slow solutions of (1) do not, in general, approximate the smoothed dynamics of (1) to any order in ϵ.

We start with the reformulation (9) of (1). Application of (6) to (9) yields

$$\begin{aligned}
\langle \frac{d}{dt} q \rangle_{\sqrt{\epsilon}} &= M^{-1} \langle p \rangle_{\sqrt{\epsilon}} \\
\langle \frac{d}{dt} p \rangle_{\sqrt{\epsilon}} &= -\langle \nabla V(q) \rangle_{\sqrt{\epsilon}} - \langle G(q)^T \lambda \rangle_{\sqrt{\epsilon}} \\
\epsilon^2 K^{-1} \langle \lambda \rangle_{\sqrt{\epsilon}} &= \langle g(q) \rangle_{\sqrt{\epsilon}}
\end{aligned} \tag{24}$$

Now (23), together with Proposition 2, implies that

$$\begin{aligned}
\langle \nabla V(q) \rangle_{\sqrt{\epsilon}} &= \langle \nabla V(q_1, q_2) \rangle_{\sqrt{\epsilon}} \\
&= \nabla V(\langle q_1 \rangle_{\sqrt{\epsilon}}, q_2) + O(\delta\, \epsilon^2) \\
&= \nabla V(\langle q \rangle_{\sqrt{\epsilon}}) + O(\delta\, \epsilon^2)
\end{aligned}$$

where we also used that $\langle q_2 \rangle_{\sqrt{\epsilon}} = q_2$. A similar statement is true for $\langle g(q) \rangle_{\sqrt{\epsilon}}$. Thus, up to terms of order $O(\delta\, \epsilon^2)$, (24) can be rewritten as

$$\begin{aligned}
\frac{d}{dt} \langle q \rangle_{\sqrt{\epsilon}} &= M^{-1} \langle p \rangle_{\sqrt{\epsilon}} \\
\frac{d}{dt} \langle p \rangle_{\sqrt{\epsilon}} &= -\nabla V(\langle q \rangle_{\sqrt{\epsilon}}) - \langle G(q)^T \lambda \rangle_{\sqrt{\epsilon}} \\
\epsilon^2 K^{-1} \langle \lambda \rangle_{\sqrt{\epsilon}} &= g(\langle q \rangle_{\sqrt{\epsilon}})
\end{aligned} \tag{25}$$

Let us assume now for a moment that, again up to terms of order $O(\delta\, \epsilon^2)$, we also have

$$\langle G(q)^T \lambda \rangle_{\sqrt{\epsilon}} = G(\langle q \rangle_{\sqrt{\epsilon}})^T \langle \lambda \rangle_{\sqrt{\epsilon}} \tag{26}$$

Then, by our assumption on the smoothing operator (6), $\langle p \rangle_{\sqrt{\epsilon}}$ must be smooth, thus $d\langle p \rangle_{\sqrt{\epsilon}}/dt$ and $\langle \lambda \rangle_{\sqrt{\epsilon}}$ have to remain bounded as $\epsilon \to 0$. Thus

$$g(\langle q \rangle_{\sqrt{\epsilon}}) = O(\epsilon^2)$$

In the limit $\epsilon \to 0$, this suggests replacing the last equation in (25) by the holonomic constraint $g(\langle q \rangle_{\sqrt{\epsilon}}) = 0$ and we obtain the constrained equations of motion

$$\begin{aligned}
\frac{d}{dt} Q &= M^{-1} P \\
\frac{d}{dt} P &= \nabla V(Q) - G(Q)^T \Lambda \\
0 &= g(Q)
\end{aligned} \tag{27}$$

Note that the new variable Λ is implicitly defined by twice differentiating the constraint $g(Q) = 0$ with respect to time. As a result we obtain

$$\Lambda \;=\; [G\,M^{-1}G^T]^{-1}\,[g_{qq}\,(M^{-1}P, M^{-1}P) - G\,M^{-1}\nabla V]$$

where, for notational convenience, we suppressed the variable Q in $V(Q)$, $G(Q)$, and the second derivative $g_{qq}(Q)$. The equations (27) constitute a constrained Hamiltonian system with Hamiltonian

$$H_\epsilon(q,p) \;=\; \frac{P^T M^{-1} P}{2} + V(Q) + g(Q)^T \Lambda$$

and constraint $g(Q) = 0$. The flow of (27) can be shown to be symplectic (in a generalized sense [9]). The solutions of (27) are also time-reversible.

The approximation (27) has been used, for example, in MD simulations to remove the bond stretching modes [17], [21]. Since $g(Q) = 0$ constrains the local variable $q_1 = g(Q)$ to its equilibrium value $q_1 = 0$, we call $g(Q) = 0$ a *hard constraint*.

Let us see now whether or not the approximation (27) yields indeed the smoothed dynamics of (1) up to terms of order $O(\epsilon^2)$. To do so, we use local coordinates and the reformulation (14) and (15) of (1). In the local coordinates (Q_1, Q_2, P_1, P_2), the constrained equations (27) are obtained by replacing the equations (14) by $Q_1 = P_1 = 0$. In terms of the variable (Q_2, P_2), this results in

$$\begin{aligned}
\frac{d}{dt} Q_2 &= B(0,Q_2)\, M^{-1} B(0,Q_2)^T P_2 \\
\frac{d}{dt} P_2 &= -\nabla_{Q_2} V(0,Q_2) - \nabla_{Q_2} \frac{P_2^T B(0,Q_2)\, M^{-1} B(0,Q_2)^T P_2}{2}
\end{aligned}$$

Since

$$\langle q_1 \rangle_{\sqrt{\epsilon}} \;=\; O(\epsilon^2)$$

and $\delta < 1$, we have

$$\langle\, \nabla_{q_2} V(q_1,q_2)\, \rangle_{\sqrt{\epsilon}} \;=\; \nabla_{q_2} V(0,q_2) + O(\epsilon^2)$$

A similar statement can be derived for

$$\langle\, \nabla_{q_2} \frac{p_2^T B M^{-1} B^T p_2}{2} \rangle_{\sqrt{\epsilon}}$$

However, because of (21) and $\langle p_1 \rangle_{\sqrt{\epsilon}} = O(\epsilon^2)$, Proposition 3 implies that

$$\langle \frac{p_1^T G(q)\, M^{-1} G(q)^T p_1}{2} \rangle_{\sqrt{\epsilon}} \;=\; \frac{m}{2}\,\delta + O(\epsilon^4)$$

where m is the number of constraints (see the Appendix). Thus the force term

$$\langle \nabla_{q_2} \frac{p_1^T G(q)\, M^{-1} G(q)^T p_1}{2} \rangle_{\sqrt{\epsilon}}$$

is not necessarily small even in the limit $\epsilon \to 0$. It has been pointed out before in the context of polymer dynamics [5] that the appropriate correction to the constrained dynamics (27) is given by the Fixman potential

$$V_F(Q) \;=\; \frac{\delta}{2} \ln\left[\det\left[G(Q)\, M^{-1} G(Q)^T\right]\right] \tag{28}$$

For a derivation of the Fixman potential see the Appendix. Note that (28) implies that (26) is true only up to terms of order $O(\delta)$.

Theorem 1. *An order $O(\epsilon^2)$ approximation of the smoothed dynamics of (1) is given by the constrained Hamiltonian equations*

$$\begin{aligned} \frac{d}{dt} Q &= M^{-1} P \\ \frac{d}{dt} P &= -\nabla V(Q) - \nabla V_F(Q) - G(Q)^T \Lambda \\ 0 &= g(Q) \end{aligned} \tag{29}$$

with Hamiltonian

$$H_e(Q,P) \;=\; \frac{P^T M^{-1} P}{2} + V(Q) + V_F(Q) + g(Q)^T \Lambda$$

Proof. (29) is equivalent to (24) up to terms of order $O(\epsilon^2)$. Standard perturbation results for differential equations (see, e.g., [18]) imply that the same is true for the solutions over bounded intervals of time.

Remarks. (i) Theorem 1 implies that the smoothed dynamics of (1) cannot, in general, be approximated by the *slow solutions* of (1) as introduced by Kreiss in [7]. One can show that, up to terms of order (ϵ^2), the slow solutions of (1) are given by the constrained equations (27) which differ from (29) by the Fixman potential (28) and thus by a term of order $O(\delta)$.

(ii) A similar result to Theorem 1 has been published before, e.g., by van Kampen [22] and Pear & Weiner [13] in the context of statistical mechanics. In [16], Rubin & Unger considered in detail the case $p_1(0) = 0$ which leads to the formulation (27) and the case $p_1(0) \neq 0$ for a single constraint; i.e. $m = 1$.

The constrained equations (29) yield satisfying results only for small enough values of ϵ^2. While, for example, $\epsilon^2 \approx 0.01$ for the force constants corresponding to bond stretching in molecular dynamics, one has to take finite size effects of ϵ into account when looking at bond-angle bending [21] where $\epsilon^2 \approx 0.1$ and $\delta \approx 0.1$. In other words, the approximation

$$\langle q_1 \rangle_{\sqrt{\epsilon}} \quad = \quad 0$$

has to be replaced by a more accurate one. This can be achieved by using $d\langle p_1 \rangle_{\sqrt{\epsilon}}/dt = O(\epsilon^2)$ to derive

$$\begin{aligned} \langle q_1 \rangle_{\sqrt{\epsilon}} \quad &= \quad -\epsilon^2 K^{-1} [\nabla_{q_1} V(0, q_2) + \nabla_{q_1} V_F(0, q_2) + \\ &\quad + \nabla_{q_1} \frac{p_2^T B(0, q_2) M^{-1} B(0, q_2)^T p_2}{2}] + O(\epsilon^4) \end{aligned}$$

Since, because of (22),

$$p_2^T B(q) M^{-1} B(q)^T p_2 \quad = \quad O(\delta) \tag{30}$$

and $V_F(Q) = O(\delta)$, an order $O(\delta\,\epsilon^2)$ estimate for $g(\langle q \rangle_{\sqrt{\epsilon}})$ is given by

$$g(Q) \quad = \quad -\epsilon^2 K^{-1} [G(Q) M^{-1} G(Q)^T]^{-1} G(Q) M^{-1} \nabla V(Q)$$

and the corresponding constrained equations of motion are now given by

$$\begin{aligned} \frac{d}{dt} Q \quad &= \quad M^{-1} P \\ \frac{d}{dt} P \quad &= \quad -\nabla V(Q) - \nabla V_F(Q) - G(Q)^T \tilde{\Lambda} \\ 0 \quad &= \quad \tilde{g}(Q) \end{aligned} \tag{31}$$

with the constraint function

$$\tilde{g} \quad := \quad g + \epsilon^2 K^{-1} [G\, M^{-1} G^T]^{-1} G\, M^{-1} \nabla V$$

In contrast to the formulation (27), the system (31) can no longer be derived from a Hamiltonian principle. However, the solutions of (31) are still time-reversible.

Upon introducing an error of order $O(\delta\,\epsilon^2)$, one can reformulate (31) as a constrained Hamiltonian system with Hamiltonian

$$H_\epsilon(Q, P) \quad = \quad \frac{P^T M^{-1} P}{2} + V(Q) + V_F(Q) + \frac{g(Q)^T K g(Q)}{2\epsilon^2} + \tilde{g}(Q)^T \hat{\Lambda}$$

where $\hat{\Lambda}$ is now of order $O(\delta)$. To see this, note that $\tilde{\Lambda}$ in (31) satisfies

$$G(Q)^T \tilde{\Lambda} \;=\; \frac{1}{\epsilon^2} G(Q)^T K^{-1} g(Q) + O(\delta)$$

Thus, since $g(Q) - \tilde{g}(Q) = O(\epsilon^2)$, $\hat{\Lambda} = O(\delta)$ and $G(Q)^T \hat{\Lambda} - \tilde{G}(Q)^T \hat{\Lambda} = O(\delta\, \epsilon^2)$.

Theorem 2. *The constrained Hamiltonian equations*

$$\begin{aligned} \frac{d}{dt} Q &= M^{-1} P \\ \frac{d}{dt} P &= -\nabla V(Q) - \nabla V_F(Q) - \frac{1}{\epsilon^2} G(Q)^T K g(Q) - \tilde{G}(Q)^T \hat{\Lambda} \\ 0 &= \tilde{g}(Q) \end{aligned} \tag{32}$$

provide an order $O(\delta\, \epsilon^2)$ approximation to the smoothed dynamics of (1).

Proof. Same as for Theorem 1.

In contrast to the constraint $g(q) = 0$, we call $\tilde{g}(q) = 0$ a *flexible constraint.*

An important aspect of Hamiltonian systems is the presence of symmetries which imply the conservation of the corresponding momentum maps (first integrals) [9]. Here we have the following

Proposition 4. *Let a Lie group Γ be a symmetry of (1) [9], i.e., $H(\gamma q, \gamma^{-T} p) = H(q,p)$ for all $\gamma \in \Gamma$, then Γ is also a symmetry of the constrained system (32), (29) respectively.*

Proof. We have to show that $H_e(\gamma Q, \gamma^{-T} P) = H_e(Q,P)$ for all $\gamma \in \Gamma$. Since $G(\gamma Q) = G(Q)\gamma^{-1}$, $\gamma^{-1} M^{-1} \gamma^T = M^{-1}$, and

$$\begin{aligned} G(\gamma Q) M^{-1} G(\gamma Q)^T &= G(Q) \gamma^{-1} M^{-1} \gamma^{-T} G(Q)^T \\ &= G(Q) M^{-1} G(Q) \end{aligned}$$

we indeed have $V_F(Q) = V_F(\gamma Q)$ and $\tilde{g}(Q) = \tilde{g}(\gamma Q)$.

Example 1. To show the effect of our two approximations (27) and (32) to the slow dynamics, we looked at a one-dimensional chain of two soft and three hard springs with both ends of the chain held fixed. The Hamiltonian,

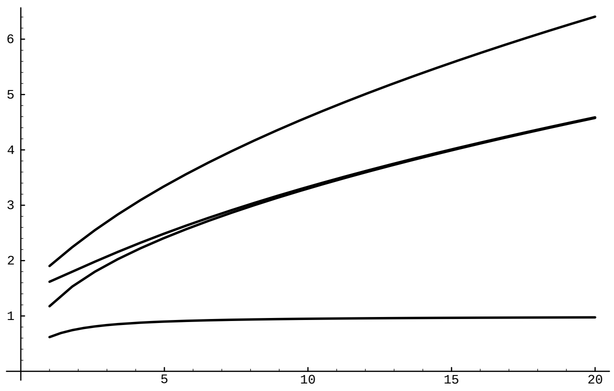

Figure 1: Natural frequencies of the unconstrained system as a function of the parameter $1/\epsilon^2$.

we used, is given by

$$\begin{aligned} H(q,p) &= \frac{p^T p}{2} + \frac{1}{2\epsilon^2}([q_1 - 1]^2 + [q_3 - q_2 - 1]^2 + [5 - q_4 - 1]^2) \\ &\quad + \frac{1}{2}([q_2 - q_1 - 1]^2 + [q_4 - q_3 - 1]^2) \end{aligned}$$

We computed the natural frequencies of the corresponding (linear) unconstrained system (1) (Fig. 1) and compared those to the ones obtained for the (linear) constrained system (27) with hard constraints (Fig. 2) and those with flexible constraints (32) (Fig. 3). Note that the smoothed dynamics is given by the smallest natural frequency of the unconstrained system. While both constrained methods correctly eliminate the three highest frequencies in the system, the low frequency component is far better approximated by the system (32) with flexible constraints. This is crucial especially for moderate values of $1/\epsilon^2$. (Note that for linear problems the Fixman potential is constant and does not need to be included into the constrained dynamics and that Theorem 2 applies with $\delta = \epsilon^2$.)

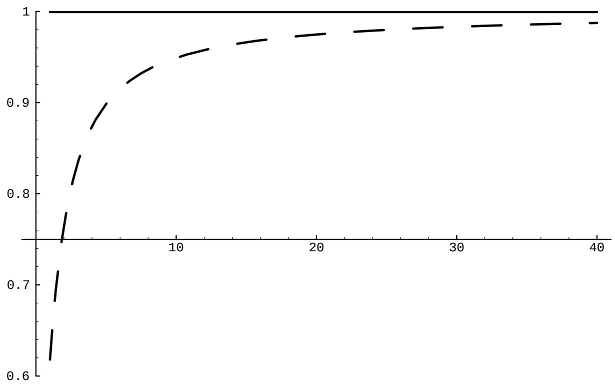

Figure 2: Natural frequency of the constrained system with hard constraints compared to the lowest frequency of the unconstrained system (dashed line) as a function of the parameter $1/\epsilon^2$.

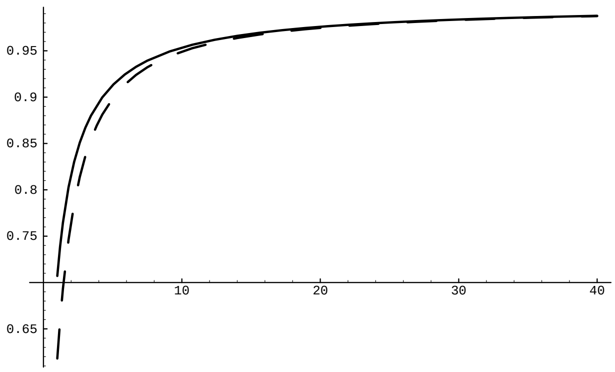

Figure 3: Natural frequency of the constrained system with flexible constraints compared to the lowest frequency in the unconstrained system (dashed line) as a function of the parameter $1/\epsilon^2$.

4 Smoothed Dynamics – Discretization

Any constrained Hamiltonian system of the form (27) can efficiently be discretized by the SHAKE extension [17]

$$\begin{aligned} Q_{k+1} &= Q_k + \Delta t\, M^{-1} P_{k+1/2} \\ P_{k+1/2} &= P_{k-1/2} - \Delta t\, [\nabla V(Q_k) + G(Q_k)^T \Lambda_k] \\ 0 &= g(Q_{k+1}) \end{aligned} \tag{33}$$

of the Verlet scheme [23] which requires now the solution of an implicit equation in the variable Λ_k. It has been shown [8] that this scheme preserves the symplectic structure [9],[19] of Hamiltonian flows, is time-reversible, and conserves first integrals related to symmetries of the system [24],[15]. Furthermore, as shown in [14], the numerical solutions can asymptotically be considered as the exact solution of a perturbed constrained Hamiltonian system.

The same scheme can also be applied to the Hamiltonian system (32) with flexible constraints. This time we obtain

$$\begin{aligned} Q_{k+1} &= Q_k + \Delta t\, M^{-1} P_{k+1/2} \\ P_{k+1/2} &= P_{k-1/2} - \Delta t\, [\nabla V(Q_k) + \frac{G(Q_k)^T K g(Q_k)}{\epsilon^2} + \tilde{G}(Q_k)^T \hat{\Lambda}_k] \\ 0 &= \tilde{g}(Q_{k+1}) \end{aligned} \tag{34}$$

Again the method is symplectic, time-reversible, and momentum conserving.

The method (34) is computational expensive. An effective implementation of (34) and the discretization of (32), (31) respectively, by less expensive methods can be found in [2]. Note that one could also discretize (32) by a proper modification of the energy-momentum methods proposed in [20].

Example 2. In this example we consider a four-bead-three-bond structure [13] where the structure is restricted to move in a finite volume by the potential

$$V_r(q) = \sum_i K_r \left(\frac{r_i}{\sigma}\right)^6$$

Here r_i denotes the distance of each of the four beads to the origin, $\sigma = 2$, and $K_r = 0.1$. We set the mass of all four beads equal to $m = 1$ and choose $r_0 = 1$ as the equilibrium bond-length and $\phi_0 = 90^o$ as the equilibrium bond-angle. For simplicity, we did not included a torsion potential. The force constant for the harmonic bond-angle bending potentials

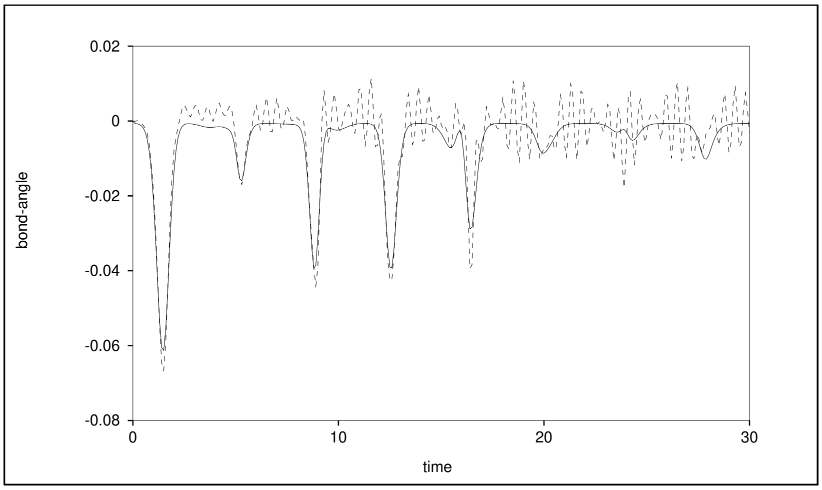

Figure 4: Trajectory of one of the bond-angles for the unconstrained formulation (dotted line) compared to the one for the formulation with hard constraints on the bond-lengths and flexible constraints on the bond-angles (solid line).

was $K_a = 60\,\mathrm{deg}^{-2}$ and $K_b = 600$ for the corresponding bond stretching potentials. Note that these values correspond to force constants typically found in molecular dynamics simulations [21].

We started the structure from its equilibrium position with the initial velocities in x-direction equal to $p_x = 1.0$. The impact of the structure clashing with the potential-wall V_r can be seen in Fig. 4. We computed the trajectory of one bond-angle (plotted as $\cos(\phi)$) for the unconstrained formulation and compared this trajectory with the one obtained by constraining the bond-lengths by hard constraints and the bond-angles by flexible constraints.

5 Appendix

The Fixman potential can easily be derived from

$$\langle\, \nabla_q \frac{p_1^T G(q)\, M^{-1} G(q)^T p_1}{2}\,\rangle_{\sqrt{\epsilon}}$$

in the following way: Let $Q(q)$ be an orthogonal matrix such that $Q(q)^T G(q)\, M^{-1} G(q)^T Q(q)$ is a diagonal matrix $D(q)$ with entries $d_{i,i}(q)$. This, together with

$$\langle p^i\, d_{i,i}\, p^i \rangle_{\sqrt{\epsilon}} \quad = \quad \langle p^i p^i \rangle_{\sqrt{\epsilon}}\, \langle d_{i,i} \rangle_{\sqrt{\epsilon}} + O(\delta\, \epsilon^2)$$

implies

$$\langle \nabla_q \frac{p_1^T G(q)\, M^{-1} G(q)^T p_1}{2} \rangle_{\sqrt{\epsilon}} \;=\; \sum_i \frac{\langle p^i p^i \rangle_{\sqrt{\epsilon}}}{2} \nabla_q d_{i,i}(\langle q \rangle_{\sqrt{\epsilon}}) + O(\delta\, \epsilon^2)$$

where p^i denotes the ith entry in the m dimensional vector $Q(q)p_1$. Since, by equipartitioning of energy (17),

$$\langle p^i p^i \rangle_{\sqrt{\epsilon}} \;=\; \frac{\delta}{d_{i,i}(\langle q \rangle_{\sqrt{\epsilon}})} + O(\delta\, \epsilon^2)$$

we obtain

$$\sum_i \frac{\langle p^i p^i \rangle_{\sqrt{\epsilon}}}{2} \nabla_q d_{i,i}(Q) \;=\; \frac{\delta}{2} \nabla_q \ln [\det\, D(Q)] + O(\delta\, \epsilon^2)$$

which, in terms of the original matrix $G(Q)M^{-1}G(Q)^T$, leads to the potential (28).

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