Computational many body quantum mechanics systems: Designing efficient numerical methods for highly-oscillatory problems

MIMOL Project

October 23, 2007

A fundamental challenge in science is the *quantitative* prediction of time-dependent nonlinear phenomena. While dynamical simulation was one of the first applications of computers, the problems treated, the methods used, and their implementation have all changed a lot. Astronomers use simulation to study the history and long term evolution of the solar system. Molecular simulations are essential for the design of new materials and for drug discovery. Simulation can replace or guide experiment, which often is difficult or even impossible to carry out as our ability to fabricate the necessary devices is limited.

At the boundaries of different modeling regimes, it is found that computations based on the fundamental laws of physics are under-resolved in the textbook sense of numerical methods. Because of the vast range of scales involved in modeling even relatively simple biological or material functions, this limitation will not be overcome by simply requiring more computing power. One therefore has to develop numerical methods which capture crucial structures even if the method is far from "converging" in the mathematical sense. In this context, we are forced increasingly to think of the numerical algorithm as a part of the modeling process itself. A major advance in this area has been the development of structure-preserving or "geometric" integrators which maintain conservation laws, dissipation rates, or other key *qualitative* features of the continuous dynamical model. Conservation of energy and momentum are fundamental for many physical models; more complicated invariants are maintained in applications such as molecular dynamics and play a key role in determining the long term stability of methods. In mechanical models (biodynamics, vehicle simulation, astrodynamics) the available structure may include constraint dynamics, actuator or thruster geometry, dissipation rates and properties determined by nonlinear forms of damping.

Features such as *symplecticity* or *time-reversibility* are now widely recognized as essential properties to preserve by numerical schemes, owing to their physical significance. This has motivated a lot of research [SSC94, HW96, HLW02] and

led to many different classes of geometric integrators (symplectic and symmetric methods, volume-preserving integrators, Lie-group methods, etc.). In practice, a few simple schemes such as the Verlet method or the Störmer methods have been used for years with great success in molecular dynamics or astronomy. However, they now need to be further improved (with respect to accuracy and computational cost) in order to fit the tremendous increase of complexity and size of the models.

1 Schrödinger equation and highly oscillatory problems

1.1 Schrödinger equation

In quantum mechanics, the evolution of particles with the time is driven by the Schrödinger equation which we write as

$$i\varepsilon \frac{\partial \psi}{\partial t} = H\psi, \tag{1}$$

where $\psi = \psi(x,t)$ is the wave function depending on the spatial variables $x = (x_1, \ldots, x_N)$ with $x_n \in \mathbb{R}^d$ (e.g., with $d = 1$ or 3 in the partition) and the time $t \in \mathbb{R}$. Here, ε is a (small) positive number representing the scaled Planck constant and i is the complex imaginary unit. The Hamiltonian operator H is written

$$H = T + V$$

with the kinetic and potential energy operators

$$T = -\sum_{j=1}^{N} \frac{\varepsilon^2}{2m_j} \frac{\partial^2}{\partial x_j^2} \quad \text{and} \quad V = V(x),$$

where $m_j > 0$ is a particle mass and where the real-valued potential V acts as a multiplication operator on ψ.

The Schödinger equation (1) exhibits a *symplectic* structure that we describe now: Let us write $\psi = v + iw$ the real and imaginary parts of $\psi \in L^2(\mathbb{R}^D, \mathbb{C})$, where $D = N \times d$. The functions v and w are thus functions in the *real* Hilbert space $L^2(\mathbb{R}^D, \mathbb{R})$. We denote the complex inner product by $\langle \cdot, \cdot \rangle$ and the real inner product by $(\cdot, \cdot)$.

As H is a real operator, Eq. (1) can be written

$$\begin{array}{rcl} \varepsilon \dot{v} & = & Hw, \\ \varepsilon \dot{w} & = & -Hv, \end{array} \tag{2}$$

or equivalently, with the canonical structure matrix

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

and the Hamiltonian

$$H(v,w) = \tfrac{1}{2}\langle \psi, H\psi\rangle = \tfrac{1}{2}(v, Hv) + \tfrac{1}{2}(w, Hw) \tag{3}$$

for $\psi = v + iw$ (we use the same symbol H as for the operator), this becomes the canonical Hamiltonian system (see eqn. (4) below)

$$\begin{pmatrix} \dot{v} \\ \dot{w} \end{pmatrix} = \varepsilon^{-1} J^{-1} \nabla H(v,w).$$

This fundamental mathematical structure of the Schrödinger equation implies in particular the conservation of the energy function (3) for the exact solution of (1). Moreover, approximating numerically the Schrödinger equation needs roughly two stages: the space approximation and the time discretization. The first step can be realized in many different ways, with the particular concern of reducing the number of degrees of freedom to make the system numerically tractable without destroying the quantum effects of the initial equation. In general, this reduction is achieved by doing a symplectic projection onto a finite dimensional submanifold of L^2 (this is the Dirac-Frenkel-McLachlan principle, see for instance [Lub05] and the reference therein). After this projection step, the obtained system of differential equations possesses a symplectic structure. Moreover, due to the semi-classical parameter ε or to small parameters in the space discretisation, these systems are naturally higly oscillatory.

After this dimension reduction, the next difficulty is to discretize these highly oscillatory hamiltonian systems with respect to the time. This is a very general and important problem in numerical analysis .

1.2 Hamiltonian systems

Like the Schrödinger equation (1), there are many different systems in nature whose evolution is accurately described by Hamiltonian systems of equations. These can be derived from a variational principle and have the following form (see (2)):

$$\begin{array}{rcrcl} \dot{p}(t) & = & -\nabla_q H(p(t),q(t)) & \in & \mathbb{R}^n \\ \dot{q}(t) & = & \nabla_p H(p(t),q(t)) & \in & \mathbb{R}^n \end{array} \tag{4}$$

with some prescribed initial values $(p(0), q(0)) = (p_0, q_0)$ and for some scalar function H, called the Hamiltonian, which in this situation is an invariant of the problem. Physically, it represents the (constant) energy of the system.

Very often, besides the Hamiltonian function, there exist other invariants for such systems (e.g., linear momentum, angular momentum, etc.): when there exist n invariants in involution, the system (4) is said to be *integrable*.

A fundamental property of Hamiltonian systems is that their exact flow (i.e., the exact solution of equations (4) is a symplectic transformation [HLW02]). In addition, integrable Hamiltonian systems behave in a very remarkable way: as a matter of fact, their invariants persist under small perturbations, as shown in

the celebrated theory of Kolmogorov, Arnold and Moser [Kol54, Arn63, Mos62, HLW02]. This behaviour motivates, in accordance with the aims of geometric integration, the introduction of *symplectic* numerical flows that approximate the exact flow when, as occurs in practice, no closed expressions can be found for the solution of (4). Symplectic integration methods preserve the symplectic structure of the Hamiltonian system and it has been shown that they also preserve a *modified* Hamiltonian function over exponentially long intervals of time. The theory sustaining this remarkable result, known as *Backward error analysis*, plays indeed a fundamental role in the analysis of geometric integrators: it basically states that the solution obtained by a numerical scheme of this kind can be considered, up to small error terms, as the *exact* solution of a modified system, close in a certain sense to the original one. This interpretation is the key to many theoretical results describing the qualitative behaviour of numerical schemes applied to Hamiltonian systems (see for instance [HLW02]).

1.3 Highly-oscillatory problems

A simple yet representative model of Hamiltonian system whose solutions are highly-oscillatory in character is given by the second-order differential system

$$\ddot{q} \quad = \quad -\nabla V(q), \tag{5}$$

where the potential $V(q)$ is a sum of potentials $V = W + U$ acting on different time-scales, with $\nabla^2 W$ positive definite and $\|\nabla^2 W\| >> \|\nabla^2 U\|$. This is typically a situation arising after the space discretisation of an infinite dimensional equation like the Schrödinger equation (1). In this case, W is a finite dimensional representation of an unbounded operator, which automatically leads to high eigenvalues ω for the Hessian matrix $\nabla^2 W$.

In order to get a bounded error propagation in the linearized equations for a given explicit numerical method, the step size h must be restricted according to

$$h\omega < C,$$

where C is a constant depending on the numerical method and ω is the largest frequency of the problem, i.e. in this situation the square root of the largest eigenvalue of $\nabla^2 W$. In applications to molecular dynamics for instance, *fast* forces deriving from W (short-range interactions) are much cheaper to evaluate than *slow* forces deriving from U (long-range interactions). In this case, it thus seems highly desirable to design numerical methods for which the number of evaluations of slow forces is not (at least not too much) affected by the presence of fast forces.

This step size restriction is intimately linked to the natural existence of two (or more) time-scales in the system. Another very undesirable consequence of these largely different time-scales is the failure of backward error analysis, for which all bounds of error terms involve the product $h\omega$. This prevents one from

drawing any conclusion from the existence of a modified system and so an alternative theory has to be proposed. Recently, Cohen, Hairer and Lubich [CHL03] have introduced the so-called *modulated Fourier expansion*, which brings new light on the behaviour of highly-oscillatory Hamiltonian systems. In their approach they consider first the (somewhat idealized) situation of two blocks of frequencies, where one corresponds to the frequency zero and the other one is scaled by a large parameter. Their contribution explains in particular the good behaviour of certain Gautschi type methods, as far as preservation of the total energy and almost invariance of oscillatory energies is concerned.

Another typical example of highly-oscillatory systems encountered in quantum dynamics are equations where the time-dependent Schrödinger equation is the model to be used, but coupled with a classical mechanical system. Assuming that the Laplacian operator has been discretized in space, one indeed gets

$$i\dot{\psi}(t) = \frac{1}{\varepsilon} H(t)\psi(t), \tag{6}$$

where $H(t)$ is a finite-dimensional matrix and where ε typically is the square-root of a mass-ratio (say electron/ion for instance) and is small ($\varepsilon \approx 10^{-2}$). Through the coupling with classical mechanics ($H(t)$ is obtained by solving some equations from classical mechanics), we are confronted once again to two different time-scale, 1 and ε. In this situation also, it is thus desirable to devise a numerical method able to advance the solution by a time-step $h > \varepsilon$. Equation (6) also appears when describing the evolution of quantum discrete-level systems in an almost-adiabatic regime. Here the challenge for the numerical is to approximate adequately both the adiabatic invariants of the problem and transition probabilities between different states.

2 Major objectives of the project

2.1 Hagedorn wave packets

In the previous collaboration [FL06], Erwan Faou and Christian Lubich derived a numerical scheme for the approximation of (1) using Gaussian wave packets in the sense of [Hel76]. In Gaussian wave packets dynamics, an approximation to the wave function $\psi(x,t)$ solution of (1) is actually sought for in the form

$$u(x,t) = e^{i\phi(t)/\varepsilon} \prod_{k=1}^{N} \varphi_k(x_k,t) \tag{7}$$

with

$$\varphi_k(x_k,t) = \exp\Big(\frac{i}{\varepsilon}\big(a_k(t)\,|x_k - q_k(t)|^2 + p_k(t)\cdot(x_k - q_k(t)) + c_k(t)\big)\Big), \tag{8}$$

where $|\cdot|$ and $\cdot$ denote the Euclidean norm and inner product on $\mathbb{R}^d$, respectively. Here, $a_k = \alpha_k + i\beta_k$ (with $\beta_k > 0$) is a complex width parameter, $c_k = \gamma_k + i\delta_k$

is a complex phase parameter, and ϕ is a real phase. The parameters $q_k \in \mathbb{R}^d$ and $p_k \in \mathbb{R}^d$ represent the position and momentum average. In [FL06], Faou & Lubich derived a numerical method to approximate the the evolution of the parameters $a_k(t)$, $c_k(t)$, $p_k(t)$ and $q_k(t)$ of the Gaussian. This scheme turns to be a symplectic integrator, that is and operator preserving the natural symplectic structure of the Gaussian wave dynamics equations inherited from the symplectic structure of the Schrödinger equation. This implies nice properties of the algorithm over very long time (in particular, the conservation of energy). Moreover, this algorithm is consistent with the semi-classical limit $\varepsilon \to 0$, where the algorithm converges towards the well known Verlet integrator widely used in classical molecular dynamics (see [HLW03]) .

One of the goal of the MIMOL project is the studying of a natural extension of this work: The approximations of the Schrödinger equation by products of complex Gaussians with polynomials. As the degrees of these polynomials increase, the corresponding submanifold of L^2 is expected to fill in the whole L^2 space, making this representation more accurate than Gaussians only.

Representing the polynomials in a basis of scaled Hermite polynomials is very appropriate in 1 space dimensions [Bil03], but in the multi-dimensional case, simply taking tensor products of Hermite polynomials (be it with a moving frame of coordinates) turns out to lead to a number of both theoretical and computational difficulties. These are overcome in an alternative extension to higher dimensions due to Hagedorn [Hag98]. While the beautiful theoretical properties of this approach are evident from [Hag98], it appears that so far they have not been put into use in computational algorithms.

This work is in progress (see [FLG07]). Erwan Faou, Christian Lubich and Vasile Gradinaru have derived a new algorithm using Hagedorn wavepackets. The goal for 2008 are the following:

- Implement the algorithm using sparse grid to be used without prohibitive cost in multidimensional problems. This extension to sparse grid is in the continuity of the work of V. Gradinaru (see [Gra07, Graar].
- Justify the algorithm: this relies on semi-classical analysis, together with space approximation, as done in [EF07]. The hope is to obtain uniform convergence estimates with respect to the semi-classical parameter ε.

2.2 Highly oscillatory problems

The main second goal of this project is to address, at least partially, some of the difficulties encountered when solving highly-oscillatory Hamiltonian systems. In particular, we will proceed along the following lines:

- For the simplest case of the highly-oscillatory nonlinear system

$$\ddot{q} + \frac{1}{\varepsilon^2}\Omega^2 q = g(q) \tag{9}$$

with $g(q) = -\nabla U(q)$ and

$$\Omega = \begin{pmatrix} 0 & 0 \\ 0 & \omega I \end{pmatrix},$$

we aim at deriving a new explicit expansion of the exact solution (and concurrently of the numerical solution) based on the explicit introduction of two time-scales (a technique usually referred to in the literature as WKB-expansions):

$$q(t) := q(t,\tau) = \sum_{j\geq 0} \omega^{-j} X^j(t,\tau) = \sum_{j\geq 0} \omega^{-j} \sum_k X_k^j(t) e^{ik\tau} \tag{10}$$

where $\tau = t\omega$ is the fast time. Though closely related to the modulated Fourier expansion, this expansion can be derived explicitly. It can further be indexed with trees in the form

$$q(t) := \sum_{u\in T} \omega^{-\rho(u)} x_u(t) e^{ik\kappa(u)} F(u)(q_0, \dot{q}_0) \tag{11}$$

where the index set T is made of appropriately chosen trees and where ρ, κ are integer functions of u, $F(u)((q_0, \dot{q}_0))$ a function based on derivatives of g and $x_u(t)$ a polynomial in t depending on u. This algebraic studying is closely related to recent works done by members of the project: see [Mur99, CM07, CFM06, HLW02].

Apart from studying the long-term behavior of the exact solution and of the numerical one with the help of (10), along the lines of Cohen, Hairer and Lubich [CHL04], we also intend to derive new order conditions -using expansion (11)- dealing with the different regimes one encounters. According to the respective sizes of h and ω^{-1}, different terms dominate the expansion, and matching them in the expansions of both the numerical and the exact solutions, lead to conditions on the method. The goal is to obtain the "classical" order conditions for various regimes: $h \leq \omega^{-1}$, $h \equiv \omega^{-1}$, $h \equiv \omega^{-1/2}$, and develop new criteria for measuring the quality of the approximation obtained by numerical methods: for instance if the averaging of both the exact solution and the numerical approximation coincide up to a certain order.

- As the next step, we will generalize the treatment to the multiple frequency case, when the matrix Ω in (9) has the form

$$\Omega = \begin{pmatrix} 0 & 0 \\ 0 & \widetilde{\Omega} \end{pmatrix}, \quad \widetilde{\Omega} = \mathrm{diag}(\omega_1, \omega_2, \ldots, \omega_k).$$

  Here the non-resonance conditions to be imposed on the numerical schemes have to be carefully analyzed.

- Almost simultaneously we will proceed to designing new methods especially tailored for the system (9) and based on the terms encountered in the WKB-expansion, in particular exponential integrators.

- We will analyze different partial differential equations which, after space discretization, result in systems of the form (9) with a specific treatment of (6).

References

[Arn63] V. I. Arnold. Small denominators and problems of stability of motion in classical and celestial mechanics. *Russian Math. Surveys*, 18:85–191, 1963.

[Bil03] G.D. Billing. The Quantum Classical Theory, *Oxford Univ. Press, 2003.* Oxford Univ. Press., 2003.

[CFM06] P. Chartier, E. Faou, and A. Murua. An algebraic approach to invariant preserving integrators: The case of quadratic and hamiltonian invariants. *Numer. Math.*, 2006.

[CHL03] D. Cohen, E. Hairer, and C. Lubich. Modulated Fourier expansions of highly oscillatory differential equations. *Foundations of Comput. Math.*, 3:327–345, 2003.

[CHL04] D. Cohen, E. Hairer, and C. Lubich. Numerical energy conservation for multi-frequency oscillatory differential equations. *Submitted*, 2004.

[CM07] P. Chartier and A. Murua. Preserving first integrals and volume forms of additively split systems. *IMA Journal of Numerical Analysis.*, 2007.

[EF07] V. Gradinaru E. Faou. Gauss-hermite wavepacket dynamics: convergence of the spectral and pseudo-spectral a pproximation,. *Submitted*, 2007.

[FL06] E. Faou and C. Lubich. A poisson integrator for gaussian wavepacket dynamics. *Computing and Visualization in Science*, 9:45–55, 2006.

[FLG07] E. Faou, C. Lubich, and V. Gradinaru. Computational many-body quantum dynamics using hagedorn wavepackets. *In preparation*, 2007.

[Gra07] V. Gradinaru. Fourier transform on sparse grids: Code design and application to the time dependent schrdinger equation on sparse grids. *Computing*, 80, 2007.

[Graar] V. Gradinaru. Strang splitting for the time dependent schrdinger equation on sparse grids. *SIAM Journal of Numerical Analysis.*, To appear.

[Hag98] G.A. Hagedorn. Raising and lowering operators for semiclassical wave packets. *Ann. Physics*, 269:77–104, 1998.

[Hel76] E.J. Heller. Time dependent variational approach to semiclassical dynamics. *J. Chem. Phys.*, 64:63–73, 1976.

[HLW02] E. Hairer, C. Lubich, and G. Wanner. *Geometric Numerical Integration. Structure-Preserving Algorithms for Ordinary Differential Equations*. Springer Series in Computational Mathematics 31. Springer, Berlin, 2002.

[HLW03] E. Hairer, C. Lubich, and G. Wanner. Geometric numerical integration illustrated by the Störmer–Verlet method. *Acta Numerica*, 12:399–450, 2003.

[HW96] E. Hairer and G. Wanner. *Solving ordinary differential equations. II: Stiff and differential-algebraic problems. 2nd rev. ed.* Springer Series in Computational Mathematics, 1996.

[Kol54] A. N. Kolmogorov. On conservation of conditionally periodic motions under small perturbations of the Hamiltonian. *Dokl. Akad. Nauk SSSR*, 98:527–530, 1954.

[Lub05] C. Lubich. On variational approximations in quantum molecular dynamics. *Math. Comp.*, 74:765–779., 2005.

[Mos62] J. Moser. On invariant curves of area-preserving mappings of an annulus. *Nachr. Akad. Wiss. Göttingen II. Math.-Phys.*, pages 1–20, 1962.

[Mur99] A. Murua. Formal series and numerical integrators, Part i: Systems of ODEs and symplectic integrators. *Applied Numerical Mathematics*, 29:221–251, 1999.

[SSC94] J. M. Sanz-Serna and M. P. Calvo. *Numerical Hamiltonian Problems*. Chapman & Hall, London, 1994.