5 Time Integration of separable Hamiltonian

Most of the model reductions from §3 yield the important subproblems (possibly as substeps of a splitting method)

$$i\dot{\psi} = -\frac{1}{2}\Delta\psi, \quad \psi(0) = \psi_0, \quad \psi \in H^2(\mathbb{R}^3).$$

After space discretization by methods from §4, the task to solve is the time integration of an ODE system

$$i\dot{y} = Ay, \quad y(0) = y_0$$

with a Hermitian matrix A of large dimension and of large norm (which typically grows with a negative power of the space discretization parameter). The solution is given by

$$y(t) = e^{-itA} y_0.$$

This is a highly oscillating function, whose efficient and numerically stable computation we discuss in the following.

<u>Remark</u>: After a pseudo-spectral space discretization with plane waves, A is diagonal, and the task trivial.

5.1 Computation of the matrix exponential

It will be seen later that it is advantageous to use a time stepping for the evaluation:

$$e^{-it_n A}\phi = \prod_{k=1}^{n} e^{-i(\Delta t_k)A}\phi, \quad \Delta t_k = t_k - t_{k-1}, \quad 0 = t_0 < t_1 < \dots < t_n$$

(with ϕ a vector).

Thus, we need to compute

$$y^{n+1} = e^{-i\Delta t A} y^n .$$

5.1.1 Chebyshev Method

Approximation by a polynomial,

$$e^{-i\Delta t A} \approx P(\Delta t A),$$

P ... a polynomial chosen a priori.

Chebyshev polynomials: For $k \in \mathbb{N}$,

$$T_k(x) := \cos(k \cdot \arccos x), \quad x \in [-1,1].$$

is a polynomial of degree k.

Recurrence relation:

$$T_{k+1}(x) = 2x\,T_k(x) - T_{k-1}(x), \quad k \geq 1.$$

$$T_0(x) \equiv 1, \quad T_1(x) = x.$$

(resulting from $\cos(n+1)\theta + \cos(n-1)\theta = 2\cos\theta\cos n\theta$).

T_k are orthogonal polynomials w.r.t. the weight function $(1-x^2)^{-1/2}$ on $[-1,1]$.

Another identity: $2T_k(x) = (x+\sqrt{x^2-1})^k + (x-\sqrt{x^2-1})^k$

Chebyshev series: For a function $f(x)$, Fourier expansion of $g(\theta) = f(\cos\theta)$ yields

$$f(x) = c_0 + 2\sum_{k=1}^{\infty} c_k T_k(x), \quad c_k = \frac{1}{\pi}\int_{-1}^{1} T_k(x) f(x) \frac{dx}{\sqrt{1-x^2}}$$

We study truncation of this series for holomorphic functions, $f(x) \approx p_m(x) = c_0 + 2\sum_{k=1}^{m} c_k T_k(x)$:

Theorem: Let $r>1$, and suppose that $f(z)$ is holomorphic in the interior of the ellipse defined by $|z+\sqrt{z^2-1}| < r$ and continuous on its closure. Then, the error of the truncated Chebyshev series is bounded by

$$|f(x) - p_m(x)| \leq 2\mu(f,r)\frac{r^{-m}}{1-1/r}, \quad 0 \leq x \leq 1,$$

with $\mu(f,r) = \frac{1}{2\pi r}\int_{|w|=r} |f(\varphi(w))|\,|dw|$, $\varphi(w) = \frac{1}{2}(w+\frac{1}{w})$.

Proof: [14, p. 87].

Theorem: The error of the Chebyshev approximation $p_{m-1}(x)$ of degree $m-1$ to the complex exponential $e^{i\omega x}$ with real ω is bounded by

$$\max_{-1\leq x\leq 1} |p_{m-1}(x) - e^{i\omega x}| \leq 4\left(e^{1-\frac{\omega^2}{4m^2}} \frac{|\omega|}{2m}\right)^m, \text{ for } m \geq |\omega|.$$

Proof: [14, p. 88].

Discussion: Thus, the error stagnates up to $m \approx |\omega|$. After this, very rapid, superlinear convergence is observed.

We arrive at the "Chebyshev Method for the Matrix Exponential Operator":

$$e^{-i\Delta t A} v \approx p_{m-1}(\Delta t A) v, \; A \text{ Hermitian, eigenvals} \in [a,b]$$

$$p_{m-1}(\Delta t A) v = e^{-i\Delta t (a+b)/2} \Big[c_0 v + \dots$$
$$\dots + 2 \sum_{k=1}^{m-1} c_k T_k \left(\frac{2}{b-a} \left(A - \frac{a+b}{2} I \right) \right) v \Big],$$

$$c_k = (-i)^k J_k\left(\Delta t \, \frac{b-a}{2}\right),$$

$$i^k J_k(\omega) = \frac{1}{\pi} \int_0^{\pi} e^{i\omega\cos\theta} \cos(k\theta)\, d\theta.$$

<u>Theorem</u>: Let A be a Hermitian matrix with all eigenvalues in the interval $[a,b]$, and v a unit vector. Then

$$\| p_{m-1}(\Delta t A)v - e^{-i\Delta t A} v \|_2 \leq$$
$$\leq 4 \left(e^{1-(\omega/2m)^2} \frac{\omega}{2m} \right)^m, \quad m \geq \omega := \frac{\Delta t (b-a)}{2}.$$

<u>Proof</u>: [14, p. 90].

<u>Discussion</u>: Thus, a wide spectrum of A necessitates small time-steps. The eigenvalues of A are usually inversely proportional to the spatial discretization parameter $\Rightarrow \Delta t \leq C m (\Delta x)^2$.

b) p_{m-1} is evaluated using "<u>Clenshaw's Algorithm</u>", based on the recurrence for the Chebyshev polynomials on p. 87.

Lanczos Method

An approach to the computation of $e^{-i(\Delta t_k)A}$ which may be an advantageous alternative to the Chebyshev method is "Lanczos Approximation", described below. A numerical comparison of methods for time integration of the separable Hamiltonian is given, for example in the context of TDDFT, in [1].

Lanczos method:

Let $A \in \mathbb{C}^{N \times N}$ be a Hermitian matrix, and $0 \neq v \in \mathbb{C}^N$.

The m-th Krylov subspace $K_m(A,v)$ w.r.t. A and v is

$K_m(A,v) = \operatorname{span}(v, Av, A^2 v, \ldots, A^{m-1} v)$.

Build an orthonormal basis of $K_m(A,v)$, starting with $v_1 = \frac{v}{\|v\|}$, and subsequent Gram-Schmidt process:

$$r_{k+1,k} \, v_{k+1} = A v_k - \sum_{j=1}^{k} (\bar{v}_j^T A v_k) \, v_j, \qquad (1)$$

$r_{k+1,k}$ s.t. $\|v_{k+1}\|_2 = 1$.

Setting $T_m := (\bar{v}_j^T A v_k)_{j,k=1}^{m}$,

note that $(T_m)_{jk} = 0$ for $j-k > 1$.

We obtain the recurrence

$$AV_m = V_m T_m + (\bar{v}_{m+1}^T A v_m) v_{m+1} e_m^T,$$

$e_m^T = (0,0,\dots,0,1)$ (m-th unit vector)

$V_m = (v_1,\dots,v_m)$, $T_m = (\gamma_{jk})$.

By orthogonality, $T_m = \bar{V}_m^T A V_m \Rightarrow$

T_m is Hermitian $\Rightarrow$ T_m is tridiagonal $\Rightarrow$

the recurrence (1) on 91 is a three-term recurrence which can be evaluated efficiently and stably [26]. (Golub & Van Loan)

<u>Theorem</u> (Optimality of Lanczos Method):

Let f be a complex-valued function defined on an interval $[a,b]$ that contains the eigenvalues of the Hermitian matrix A, and let v be a vector of unit norm. Then, the error of the Lanczos approximation to $f(A)v$ is bounded by ($f(A) = U \operatorname{diag}(f(\lambda_j)) \bar{U}^T$ for $A = U \operatorname{diag}(\lambda_j) \bar{U}^T$)

$$\| V_m f(T_m) \underset{\substack{\shortparallel \\ (1,0,\dots,0)^T}}{e_1} - f(A)v \|_2 \leq 2 \inf_{p_{m-1}} \max_{x \in [a,b]} |p_{m-1}(x) - f(x)|$$

with the infimum taken over all polynomials of degree $\leq m-1$. <u>Proof</u>: [14, p. 95]

Combining the last result with the estimate for the Chebyshev approximation on p. 89, which serves as an upper bound on the infimum in the last Theorem, we can sum up:

<u>Theorem</u>: Let A be an Hermitian matrix all of whose eigenvalues are in the interval $[a, b]$, and let v be a vector of unit Euclidean norm. Then, the error of the Lanczos method is bounded by

$$\| V_m e^{-i\Delta t T_m} e_1 - e^{-i\Delta t A} v \|_2 \leq 8 \left(e^{1-\omega^2/4m^2} \frac{\omega}{2m} \right)^m, \quad m \geq \omega,$$

with $\omega = \Delta t\,(b-a)/2$.

<u>Remark</u>: The step-size restriction $\Delta t \leq C_m (\Delta x)^2$ also applies for this result!

<u>Remark</u>: Further methods to evaluate the matrix exponential in question are discussed in [17] for instance.

Methods for a time-dependent Hamiltonian

Consider a Schrödinger equation after (any suitable) space discretization

$$i\dot{\psi} = H(t)\psi, \quad \psi(0) = \psi_0,$$

with a Hamiltonian explicitly depending on time.

A popular integrator is the "<u>implicit midpoint rule</u>" or "<u>Crank-Nicholson method</u>"

$$i\,\frac{\psi_{n+1}-\psi_n}{\Delta t} = H(t_{n+1/2})\,\frac{\psi_{n+1}+\psi_n}{2}, \quad n = 0, 1, \ldots$$

This method has a <u>unitary propagator</u>:

$$\psi_{n+1} = r(-i\Delta t H(t_{n+1/2}))\psi_n, \quad r(z) = \frac{1+\frac{z}{2}}{1-\frac{z}{2}}, \quad |\psi_{n+1}| = |\psi_n|.$$

This is an important geometric property the numerical method shares with the exact flow. Thus, the L^2-norm is preserved,

$$\|\psi_{n+1}\|_{L^2} = \|\psi_n\|_{L^2} = \ldots = \|\psi_0\|.$$

It is also <u>time reversible</u>, i.e. invariant under exchange $n \longleftrightarrow n+1$ and $\Delta t \longleftrightarrow -\Delta t$, or equivalently,

$$r(z)^{-1} = r(-z).$$

It shares this property with the exponential function e^z.

It is well-known that this method has the classical convergence order two. The classical error analysis cannot be applied in the presence of an unbounded operator, however. A careful analysis shows that second order convergence also holds for unbounded $H(t)$ if $\dot{H}, \ddot{H}, \psi^{(3)}$ are bounded [11]. An alternative is the "<u>exponential midpoint rule</u>"

$$\psi_{n+1} = \exp(-i \Delta t\, H(t_{n+1/2}))\, \psi_n$$

Computation of the matrix exponential is realized by the methods discussed earlier. The exponential midpoint rule has a unitary propagator and is time-reversible. A second-order error bound can be shown for $\psi \in H^1$ and bounded potential [40].

Higher-order Magnus integrators:

Consider $i\dot{\psi} = H(t)\psi$, $\psi(0) = \psi_0$ (1)

In the scalar case, or when $H(t)$ and $\int_0^t H(\tau)\,d\tau$ commute, the solution can be represented as

$$\psi(t) = \exp\left(-i\int_0^t H(\tau)\,d\tau\right)\psi_0 .$$

In general, we wish to find a matrix $\Omega(t)$ s.t.

$$\psi(t) = \exp(\Omega(t))\psi_0$$

solves $\dot{\psi} = A(t)\psi(t)$

It can be shown [8] that this implies the nonlinear differential equation

$$\dot{\Omega} = A(t) - \frac{1}{2}[\Omega, A(t)] + \frac{1}{12}[\Omega,[\Omega, A(t)]] + \ldots$$

Integration and application of Picard iteration to approximate the nonlinear problem yields

$$\Omega(t) = \int_0^t A(\tau)\,d\tau - \frac{1}{2}\int_0^t \left[\int_0^\tau A(\sigma)\,d\sigma, A(\tau)\right] d\tau +$$
$$+ \frac{1}{4}\int_0^t \left[\int_0^\tau \left[\int_0^\sigma A(\mu)\,d\mu, A(\sigma)\right] d\sigma, A(\tau)\right] d\tau +$$
$$+ \frac{1}{12}\int_0^t \left[\int_0^\tau A(\sigma)\,d\sigma, \left[\int_0^\tau A(\mu)\,d\mu, A(\tau)\right]\right] d\tau + \ldots \quad (2)$$

This is called the "Magnus expansion"

Practical evaluation of the expression for $\Omega(t)$ implies two approximations:

1) Truncation of the infinite series
2) Numerical quadrature

Thus, a whole plethora of Magnus integrators results.

•) Truncation after the first term, quadrature by midpoint rule. This yields

$$\Psi_{n+1} = \exp(-i \Delta t\, H(t_{n+1/2}))\, \Psi_n ,$$

the <u>exponential midpoint rule</u>, a second order approximation [8]

•) Truncation of (2) on p. 96 after the four terms given, two-stage Gauss quadrature.

This yields

$$\Psi_{n+1} = \exp\left(-i \frac{\Delta t}{2}(H_1 + H_2) - \frac{\sqrt{3}(\Delta t)^2}{12}[H_2, H_1]\right) \Psi_n ,$$

$$H_1 := H(t_n + c_1 \Delta t), \quad H_2 := H(t_n + c_2 \Delta t),$$

$$c_{1,2} = \frac{1}{2} \pm \frac{\sqrt{3}}{6} \quad \text{--- Gauß nodes,}$$

a fourth order approximation [8].

For quantum dynamics, the classical order four can be established for $\Psi \in H^3$ and under further assumptions on the regularity of V [40].

Furthermore, the propagators are <u>unitary</u> and the methods <u>time-reversible</u>.