ESAIM: M2AN 53 (2019) 2109–2119
https://doi.org/10.1051/m2an/2019033

ESAIM: Mathematical Modelling and Numerical Analysis
www.esaim-m2an.org

CONVERGENCE OF EXPONENTIAL LAWSON-MULTISTEP METHODS FOR THE MCTDHF EQUATIONS

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Abstract. We consider exponential Lawson multistep methods for the time integration of the equations of motion associated with the multi-configuration time-dependent Hartree–Fock (MCTDHF) approximation for high-dimensional quantum dynamics. These provide high-order approximations at a minimum of evaluations of the computationally expensive nonlocal potential terms, and have been found to enable stable long-time integration. In this work, we prove convergence of the numerical approximation on finite time intervals under minimal regularity assumptions on the exact solution. A numerical illustration shows adaptive time propagation based on our methods.

Mathematics Subject Classification. 65L05, 65L06, 65M12, 65M15, 65M20, 81-08.

Received March 30, 2018. Accepted April 25, 2019.

1. Introduction and overview

We study time integration methods for nonlinear Schrödinger equations of the type

$$\partial_t \psi(t) = A\psi(t) + B(\psi(t)) = -\mathrm{i}\hat{H}(\psi(t)), \quad t > t_0, \tag{1.1}$$

on the Hilbert space L^2. Here, $A : \mathcal{D} \subseteq L^2 \to L^2$ is a self-adjoint differential operator and B a generally unbounded nonlinear operator. We will denote the flow of $-\mathrm{i}\hat{H}$ by $\mathcal{E}_{-\mathrm{i}\hat{H}}$, that is, $\psi(t) = \mathcal{E}_{-\mathrm{i}\hat{H}}(t, \psi_0)$ is the solution of $\partial_t \psi = -\mathrm{i}\hat{H}(\psi),\ \psi(0) = \psi_0$, and similarly for $\mathcal{E}_A$ and $\mathcal{E}_B$.

Equations of this type commonly arise from model reductions of high-dimensional quantum dynamical systems serving to make many-particle quantum simulations computationally tractable. Examples of such model reductions are the Gross–Pitaevskii equation (GPE) for Bose–Einstein condensates (BEC) [41], which constitutes a meanfield theory for ultracold dilute bosonic gases, the multiconfigurational time-dependent Hartree for bosons (MCTDHB) method for ultracold bosonic atoms [1], the multiconfigurational time-dependent Hartree–Fock (MCTDHF) method [27,32,48] and its most current extension, the time-dependent complete-active-space self-consistent-field (TD-CASSCF) method for electron dynamics in atoms and small molecules [44], and time-dependent density functional theory (TDDFT) for extended systems such as large molecules, nanostructures

Keywords and phrases. Multi-configuration time-dependent Hartree–Fock method, exponential Lawson multistep methods, stability, local error, convergence.

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and solid-state systems [45]. All of these *ab initio* methods imply (systems of) nonlinear Schrödinger equations for the orbitals which are highly sensitive to the accuracy of numerical integration, and whose propagation constitutes a major challenge, especially for strongly correlated systems. Moreover, they share the property that the nonlinear operator B is expensive to evaluate and thus, it is of importance that a numerical time propagator requires a minimum of evaluations of this operator, but still retains accuracy and stability, also in long-term integration.

Different approaches to the numerical solution of quantum dynamical systems have been discussed in the literature, see for instance the comprehensive overview [2]. While for the *ab initio* solution of the linear two-particle Schrödinger equation the short iterative Lanczos algorithm has proven to be very efficient [20], it is not suitable for nonlinear orbital equations especially in the presence of strong nonlinearity. Often Runge–Kutta algorithms are used within multiconfigurational methods because of their simplicity and broad applicability [12, 44]. However, explicit finite difference methods like Runge–Kutta or linear multistep methods typically suffer from instabilities unless time-steps are very small, while implicit methods like Crank–Nicolson have attractive stability and conservation properties, but the computational effort for the solution of the nonlinear equations is a serious drawback. Semi-implicit relaxation methods introduced in [43], which only treat the linear term implicitly, share the conservation properties if only a cubic nonlinearity is present and the kinetic part is treated implicitly, but they are still computationally expensive and suffer from stability limitations, see [2]. Semi-implicit finite difference schemes lose most of the desired properties. A relaxation method based on a finite element spatial discretization was recently found to perform well in [24] for problems with low spatial regularity. Extensive numerical comparisons in the literature show the accuracy and efficiency of splitting methods for various quantum mechanical models under a number of different spatial discretizations, see for instance [8, 16].

Our focus is on the equations of motion associated with the MCTDHF approximation to the multi-particle electronic Schrödinger equation, where the key issue is the high computational effort for the evaluation of the nonlocal (integral) operator B. Thus, in the choice of the most appropriate integrator, we emphasize a minimal number of evaluations of B for a given order and disregard the effort for the propagation of A, which can commonly be realized at essentially the cost of two (cheap) transforms between real and frequency space. It turns out that splitting methods, whose convergence for the MCTDHF equations has been analyzed in [30, 32], require a prohibitive number of evaluations of B to obtain a high-order approximation, whence in spite of their favorable properties, splitting methods are computationally too expensive for our purpose. A separate treatment of the two vector fields has been used, *e.g.* in [23] where the orbital equations of motion are split from the equations of motion for the configuration amplitudes and a matrix exponentiation algorithm is applied. The approach that we investigate in this paper is also based on splitting of the vector fields in (1.1). We will study *exponential Lawson multistep methods*, where a transformation is used to cope with the stiff kinetic operator, and subsequently the resulting smoother problem is solved by an Adams multistep method. It is demonstrated in the forthcoming study [3] that for the MCTDHF equations we consider here, exponential Lawson multistep methods show clear advantages over the competing methods (like explicit Runge–Kutta, linear multistep, exponential Runge–Kutta or splitting methods) in that they enable stable long-time integration when used in a predictor-corrector framework, they provide cheap local error estimators to serve as the basis for adaptive step-size choice, and in this setting only require two evaluations of B in each step for approximation of arbitrarily high order. We show an illustration of an adaptive simulation based on such a strategy for error control in Section 6.

In this paper, we give a theoretical error analysis for Adams–Lawson multistep methods on finite time intervals, to serve as a basis for their reliable application to the MCTDHF equations. In Section 2, we introduce the numerical methods, in Section 3 we briefly describe the MCTDHF method. Section 4 lists analytical prerequisites used in the proof, which is carried out in Section 5. A numerical illustration is given in Section 6.

2. Lawson methods

In Lawson methods, the equation (1.1) is transformed prior to the numerical integration by the substitution

$$\psi(t) \to \mathrm{e}^{-tA}\psi(t). \tag{2.1}$$

To the resulting equation

$$\psi'(t) = \mathrm{e}^{-tA} B(\mathrm{e}^{tA}\psi(t)) \equiv F(\psi(t)), \tag{2.2}$$

any appropriate time-stepping scheme can be applied. The main advantage lies in the fact that the dynamics associated with the non-smooth operator A is separated by the transformation, which can be realized cheaply for example in frequency space, while the problem subjected to the time-stepping scheme is smoother, thus allowing for larger time-steps. This transformation was first introduced in [34] for ordinary differential equations, where Runge–Kutta methods were applied to the transformed problem and a classical non-stiff error analysis was applied to the resulting equation. In modern theoretical physics, the method was made known by the two Ph.D. theses [15, 17], where the classical explicit fourth order Runge–Kutta method was employed for the solution of the transformed problem. Explicit Lawson multistep methods were introduced in [33] together with an extension to improve the accuracy. Hult [26] compares explicit Lawson Runge–Kutta methods for the cubic nonlinear Schrödinger equation with methods like "Blow–Wood" or splitting, with the best results for the Lawson approach. Adaptive time-stepping has been considered for these methods in a number of papers. Balac and Mahé [7] compares error estimates based on mesh halving to the cheaper embedded Runge–Kutta methods [19], while in [14], projected Lawson Runge–Kutta methods are compared to splittings with embedded error estimators [31], with an ambivalent picture favoring Lawson methods for higher orders (the splitting methods considered are not optimal in terms of work/precision, *cf.* [4]). The method from [7] has been implemented and applied to the simulation of light-wave propagation in optical fibers in [5]. A similar approach is adopted in [46], where comparisons are made also for the Burgers equation with shocks. Here, an adaptive approach is favorable for the nonlinear Schrödinger equation, and parallelization issues are also discussed. Balac *et al.* [6] studies mathematical features of the Lawson explicit fourth order Runge–Kutta method from [26] and gives an error analysis, which is however based on the assumption of the regularity of the numerical solution. Comparisons are given with Strang splitting, where however the propagation of the nonlinear part is not realized favorably. Adaptivity for embedded Lawson Runge–Kutta methods shows an advantage for one of the investigated nonlinear Schrödinger equations. Besse *et al.* [11] studies exponential time propagation methods for the rotational Gross–Pitaevskii equation by comparing exponential Runge–Kutta methods with Lawson and splitting methods, with no clear advantage for either approach. An error analysis given there only considers the transformed problem, without taking into account the stiffness hidden in the Lawson transformation. The very recent work [25] gives a convergence proof of Lawson Runge–Kutta methods in the stiff case, however under the assumption that the operator B is smooth, which is not the case in the MCTDHF equations we are considering.

In a different interpretation of exponential Lawson multistep methods, the variation of constant formula (VOC) is used to express the solution of (1.1) for a time-step $t_n \to t_{n+1} = t_n + h$ as

$$u(t_n + h) = \mathrm{e}^{hA} u_n + \mathrm{e}^{hA} \int_0^h \mathrm{e}^{-\tau A} B(u(t_n + \tau))\, \mathrm{d}\tau, \tag{2.3}$$

where the whole integrand is replaced by a polynomial interpolant. We are mainly interested in multistep methods, as evaluations of B are prohibitively expensive, so exponential Lawson multistep methods of Adams type are constructed by replacing the integrand by an interpolation polynomial of degree $k-1$ in the explicit Adams–Bashforth methods and degree k for implicit Adams–Moulton methods, interpolating the points

$$(-(k-1)h, \mathrm{e}^{(k-1)hA} B(u_{n-k+1})), \ldots, (-h, \mathrm{e}^{hA} B(u_{n-1})), (0, B(u_n)), \ [(h, \mathrm{e}^{-hA} B(u_{n+1}))].$$

The last point in square brackets is used for implicit methods, while otherwise the methods are explicit.

For our purpose, exponential Lawson multistep methods are cast as a finite difference method

$$u_{n+1} = \mathrm{e}^{hA} u_n + h \sum_{j=1}^{k} \beta_j \mathrm{e}^{(k-j+1)hA} B(u_{n-k+j}) \ [+h\beta_{k+1} B(u_{n+1})], \quad n \geq k, \tag{2.4}$$

where the last term appears in implicit Adams–Moulton type schemes and the values $u_0, \ldots, u_{k-1}$ are obtained by a suitable auxiliary scheme ensuring order k. This is equivalent to the application of the corresponding linear

multistep method to the transformed equation (2.2). We demonstrate in [3] that the best approach for our goal is to use exponential Lawson multistep methods in a predictor-corrector implementation, which is shown to enhance stability of the numerical method for long-time integration, and also provides a local error estimator for adaptive time-stepping at no additional cost. The efficiency of the time-discretization can be improved if high-order time propagators are employed. In the multistep approach, this does not imply additional computational cost if no memory limitations have to be taken into account. In Section 6 we demonstrate an adaptive strategy based on a predictor-corrector method of order six.

3. The MCTDHF method

Subsequently, we will analyze the convergence of exponential Lawson multistep methods for the time integration of the equations of motion associated with the multi-configuration time-dependent Hartree–Fock method (MCTDHF) for the approximate solution of the time-dependent multi-electron Schrödinger equation for f particles

$$\partial_t \psi = -\mathrm{i}\, H\psi, \tag{3.1a}$$

where the complex-valued wave function $\psi = \psi(x_1, \dots, x_f, t)$ explicitly depends on time t and, in the case considered here, the positions $x_1, \dots, x_f \in \mathbb{R}^3$ of electrons in an atom or molecule. The Hamiltonian H is time-dependent and has the form

$$-\mathrm{i}\, H(t) := \mathrm{i} \sum_{k=1}^{f} \left(\frac{1}{2}\Delta^{(k)} + U(x_k) + \sum_{l<k} V(x_k - x_l) \right) + \mathrm{i} V_{\mathrm{ext}}(x_1, \dots, x_f, t) =: T + W(x_1, \dots, x_f, t), \tag{3.1b}$$

where

$$T := \frac{\mathrm{i}}{2} \sum_{k=1}^{f} \Delta^{(k)} \tag{3.1c}$$

$$U(x) := \frac{Z}{|x|}, \quad Z \in \mathbb{N}, \tag{3.1d}$$

$$V(x-y) := -\frac{1}{|x-y|}. \tag{3.1e}$$

$V_{\mathrm{ext}}(x_1, \dots, x_f, t)$ is a smooth time-dependent function, and $\Delta^{(k)}$ is the Laplace operator with respect to x_k only. For the purpose of this analysis, we will disregard the nuclear attractive force U and only consider models of a free electron gas (or *jellium*) as for instance in [38–40], and also neglect the external potential V_{ext}, which is not critical if it is sufficiently smooth.

In MCTDHF as put forward for example in [13, 40, 47, 48], the multi-electron wave function ψ from (3.1) is approximated in the manifold of functions $\mathcal{M}$ satisfying the ansatz

$$u = \sum_{(j_1, \dots, j_f)} a_{j_1, \dots, j_f}(t) \phi_{j_1}(x_1, t) \cdots \phi_{j_f}(x_f, t) =: \sum_{J} a_J(t) \Phi_J(x, t), \tag{3.2}$$

where ϕ_j depend on the coordinates of one particle only and are referred to as *single-particle functions* or *orbitals*. Using (3.2) for the *electronic* Schrödinger equation, the Pauli principle implies that only solutions u are considered which are antisymmetric under exchange of any two of their arguments x_j, x_k. This assumption is particular to the MCTDHF approach, as compared to the multi-configuration time-dependent Hartree method (MCTDH) proposed in [9, 10, 36, 37] for quantum molecular dynamics. Antisymmetry reduces the number of equations considerably. Particularly, the assumption implies antisymmetry in the coefficients a_J. Formally, multi-indices $J = (j_1, \dots, j_f)$ vary for $j_k = 1, \dots, N,\ k = 1, \dots, f$. Due to the simplifications resulting from the antisymmetry assumption, only $\binom{N}{f}$ equations for a_J have to be solved in the actual computations, however.

In the case of the electronic Schrödinger equation which we are focussing on here, we further have to take into account electron spin. However, it was explained for example in [28] that this does not change our mathematical considerations concerning the equations of motion associated with MCTDHF as it only augments the system of equations without a change in their structure. Consequently, we ignore spin and concentrate on the representation (3.2). To define the MCTDHF method, we will resort to the Hilbert spaces L^2 with inner product $\langle u|v\rangle = \int \bar{u}v \,\mathrm{d}x$, where x comprises all spatial coordinates that both u and v depend on.

The Dirac–Frenkel variational principle [18,21] is used to derive differential equations for the coefficients a_J and the single-particle functions ϕ_j in (3.2). Thus, for $u \in \mathcal{M}$ we require

$$\left\langle \delta u \middle| (\mathrm{i}\partial_t - H)u \right\rangle = 0, \tag{3.3}$$

where δu varies in the tangent space $\mathcal{T}_u\mathcal{M}$ of $\mathcal{M}$ at u.

It was shown in [29] that the set $\mathcal{M}$ in conjunction with a full-rank-condition for the *density matrix* ρ defined in (3.5d) below can be endowed with the structure of a manifold, justifying the application of the variational principle as explained above. We do not give details here, but henceforth consider $\mathcal{M}$ a manifold under the tacit assumption that ρ is nonsingular.

In order to define a unique solution of (3.3), we impose additional constraints,

$$\left\langle \phi_j \middle| \phi_k \right\rangle = \delta_{j,k}, \quad t \geq 0, \tag{3.4a}$$

$$\left\langle \phi_j \middle| \partial_t \phi_k \right\rangle = -\mathrm{i} \left\langle \phi_j | T\phi_k \right\rangle. \tag{3.4b}$$

The variational principle (3.3) and the additional restrictions (3.4) finally yield equations of motion for the coefficients and single-particle functions in (3.2):

$$\mathrm{i}\frac{\mathrm{d}a_J}{\mathrm{d}t} = \sum_K \left\langle \Phi_J \middle| W\Phi_K \right\rangle a_K, \quad \forall J, \tag{3.5a}$$

$$\mathrm{i}\partial_t \phi_j = -\frac{1}{2}\Delta^{(j)}\phi_j + (I-P)\sum_{k=1}^{N}\sum_{l=1}^{N} \rho^{-1}_{j,l}\overline{W}_{l,k}\phi_k, \quad j = 1,\ldots,N, \tag{3.5b}$$

where

$$\psi_j := \langle \phi_j | u \rangle, \tag{3.5c}$$

$$\rho_{j,l} := \langle \psi_j | \psi_l \rangle, \tag{3.5d}$$

$$\overline{W}_{j,l} := \langle \psi_j | W\psi_l \rangle, \tag{3.5e}$$

$\Delta^{(j)}$ is the Laplacian with respect to the coordinates of the jth particle, and P is the orthogonal projector onto the space spanned by the functions ϕ_j. We will henceforth denote

$$A := \frac{\mathrm{i}}{2}(0, \Delta^{(1)}, \ldots, 0, \Delta^{(f)})^T \tag{3.5f}$$

and

$$B = B(a, \phi) \tag{3.5g}$$

the vector of the components associated with the potential to correspond with (1.1), where we denote the coefficient tensor by $a = (a_J)$ and the vector of orbitals by $\phi = (\phi_j)$. Clearly, the high-dimensional integrals appearing in (3.5) imply a huge computational effort for each evaluation, whence the number of evaluations of B should be kept to a minimum as detailed above.

Remark 3.1. Note that under the full-rank condition on ρ, there is a one-to-one correspondence between the coefficients a_J and orbitals ϕ_j on the one hand and the approximate wavefunction u from (3.2). One implication is trivial, and the other readily follows from the fact that under the gauge conditions (3.4), the equations of motion (3.5) which define a unique solution can be derived [30].

We will therefore tacitly use u and (a, ϕ) synonymously henceforth. Due to the product structure of u, it is also clear that norms are equivalent for both interpretations.

4. Prerequisites

In order to prove convergence of exponential Lawson multistep methods for the MCTDHF equations, we will resort to a number of results, which are either well established facts about multistep methods which can be found for instance in the monographs [22, 42] or have been derived for the analysis of splitting methods for the MCTDHF equations in earlier work by the author [30, 32]. We will analyze the convergence within the framework of multistep methods when applied to the transformed problem (2.2).

4.1. Stability and convergence of multistep methods

An Adams-type multistep method defined by coefficients $\beta_1, \dots, \beta_k, [\beta_{k+1}]$ (see (2.4)) has formal order k in the explicit and order $k+1$ in the implicit case, where the order is defined in terms of the asymptotics for $h \to 0$ of the local error as given in Table III.2.1 of [22] and the definition of the local error $u(t_n) - \tilde{u}_n$ ($\tilde{u}_n$ is the multistep approximation with exact starting values $\tilde{u}_\ell = u(t_\ell),\ \ell = 0, \dots, k-1$) is given in Definition III.2.1 of [22], see also equation (11.47) of [42]. The time integrator is said to be *stable* if the *root condition* is satisfied (also referred to as *zero-stability* or *D-stability*, see [22], Def. III.3.2 or [42], Def. 11.19). This means that all the roots of the method's first characteristic polynomial $\sum_{j=1}^{k[k+1]} \beta_j z^j$ lie in the unit circle, and the roots of modulus 1 are simple. This condition is known to be satisfied for Adams-type methods of any order [22]. From *consistency* (the local error tends to 0 as $h \to 0$) and stability, convergence of the numerical approximation follows, see Theorem III.4.5 of [22] and Theorem 11.5 of [42]:

Theorem 4.1. *Assume that for a linear multistep method of order p the starting values (obtained by a suitable one-step method) satisfy*

$$\|u_n - u(t_n)\| = O(h^p), \quad n = 0, \dots, k-1.$$

Furthermore, assume the right-hand side F in an evolution equation (2.2) to be continuous and locally Lipschitz (see [22], (4.2) on p. 391). If the problem is solved by a stable multistep method of order p and the solution is sufficiently smooth ($u \in C^{p+1}$), then the method is convergent of order p, i.e.

$$\|u_j - u(t_j)\| = O(h^p), \qquad j \geq 0.$$

If $u \in C^{q+1}$ only, with $q < p$, then

$$\|u_j - u(t_j)\| = O(h^q), \qquad j \geq 0.$$

4.2. Sobolev estimates for the MCTDHF equations

In order to analyse the convergence of exponential Lawson multistep methods applied to the MCTDHF equations (3.5), we will verify the assumptions of Theorem 4.1. To this end, we will resort to a number of estimates which have been derived earlier for the purpose of the analysis of splitting methods applied to the same problem. Firstly, we note bounds on the flows of the sub-operators which follow from (3.5), (3.8) of [30] and the proof of Theorem 3.2 of [32]. These are defined on Sobolev spaces H^k with associated norms $\|\cdot\|_{H^k}$, which are comprised of all functions in L^2 with k weak derivatives:

Lemma 4.2. *For the flows defined by the operators in (3.5), the following estimates hold:*

$$\begin{aligned}
&\|\mathcal{E}_A(t,u)\|_{H^m} = \|u\|_{H^m}, && m \geq 0,\\
&\|B'(u)\|_{L^2 \leftarrow L^2} \leq C\|u\|_{L^2}, && C = C(\|u\|_{H^1}),\\
&\|B'(u)\|_{H^m \leftarrow H^m} \leq C\|u\|_{H^m}, && C = C(\|u\|_{H^m}), \quad m \geq 1,\\
&\|\mathcal{E}_B(t,u) - \mathcal{E}_B(t,\tilde{u})\|_{H^m} \leq \mathrm{e}^{C\,t}\|u-\tilde{u}\|_{H^m}, && C = C(\|u\|_{H^m}, \|\tilde{u}\|_{H^m}), \quad m \geq 1.\\
&\|B''(u)\|_{L^2 \leftarrow (L^2 \times L^2)} \leq C(\|u\|_{H^1}),\\
&\|B''(u)\|_{H^m \leftarrow (H^m \times H^m)} \leq C(\|u\|_{H^m}), && m \geq 1,
\end{aligned}$$

where C is a generic constant (which is not the same in general in the different bounds) and B' denotes the Fréchet derivative with respect to u (or synonymously, (a,ϕ)). C depends on the respective norms in a generally nonlinear way.

All commutator bounds given in Lemma 4.3 have been derived in [30,32] or can be concluded analogously in a straightforward manner. Here, $\mathrm{ad}_A^1(B)u = [A,B](u) = AB(u) - B'(u)Au$ denotes the commutator of the two vector fields, and iterated commutators are defined by $\mathrm{ad}_A^{j+1}(B)u = [A, \mathrm{ad}_A^j(B)]u,\ j \geq 1$.

Lemma 4.3. *The operators A and B associated with the MCTDHF equations (3.5) satisfy the iterated commutator bounds*

$$\begin{aligned}
&\left\|\mathrm{ad}_A(B)\,u\right\|_{L^2} \leq C(\|u\|_{H^1}),\\
&\left\|\mathrm{ad}_A(B)\,u\right\|_{H^1} \leq C(\|u\|_{H^2}),\\
&\left\|\mathrm{ad}_A^2(B)\,u\right\|_{L^2} \leq C(\|u\|_{H^2}),\\
&\left\|\mathrm{ad}_A^2(B)\,u\right\|_{H^1} \leq C(\|u\|_{H^3}),\\
&\left\|\mathrm{ad}_A^{j+2}(B)\,u\right\|_{H^k} \leq C(\|u\|_{H^{k+2\,j+1}}), \quad j \geq 1, \quad k = 0,1.
\end{aligned}$$

Finally, we will also require Lipschitz bounds for the right-hand side of (3.5) in Sobolev spaces. These are again given in the proof of Theorem 3.1 of [30].

Lemma 4.4. *In a Sobolev space S, $S \in \{L^2, H^1, H^2\}$, the following estimates hold for the data a, $\tilde{a}$, ϕ, $\tilde{\phi}$ appearing in the MCTDHF equations (3.5):*

$$\|B(a,\phi) - B(\tilde{a},\tilde{\phi})\|_S \leq C_1\|(a,\phi) - (\tilde{a},\tilde{\phi})\|_S, \quad C_1 = C_1(\|\phi\|_{H^1}, \|\tilde{\phi}\|_{H^1}, \|a\|, \|\tilde{a}\|).$$

5. Convergence proof

The proof proceeds similarly as in [30,32,35], a two-stage ("boot-strapping") convergence proof is adopted, since the discrete evolution operator is stable only under the condition that the numerical solution is in H^1. Thus, we first show convergence in H^1, and subsequently we analyse convergence in L^2. To this end, we will verify the assumptions of Theorem 4.1 in the respective spaces, and resort to the estimates from Section 4.2 for this purpose.

(1) *Lipschitz condition in H^1.* The Lipschitz condition for the right-hand side F in (2.2) defined by (3.5) follows from Lemma 4.4.

(2) *Local error bound in H^1.* In order to estimate the local error, we resort to Theorem 4.1. Thus we verify the regularity requirements for $u \in C^{q+1}$, or equivalently, $F \in C^q$. In that case, the order of the local error is $q+1$. We compute the time derivatives of F, and obtain

$$\begin{aligned}
F(u(t)) &= \mathrm{e}^{-tA}B(\mathrm{e}^{tA}u(t)),\\
\partial_t F(u(t)) &= -\mathrm{e}^{-tA}[A,B](\mathrm{e}^{tA}u(t)) + \mathrm{e}^{-tA}B'(\mathrm{e}^{tA}u(t))(B(\mathrm{e}^{tA}u(t))),
\end{aligned}$$

$$\begin{aligned}\partial_{tt}F(u(t)) &= \mathrm{e}^{-tA}[A,[A,B]](\mathrm{e}^{tA}u(t)) - 2\mathrm{e}^{-tA}AB'(\mathrm{e}^{tA}u(t))B(\mathrm{e}^{tA}u(t))\\ &\quad + \mathrm{e}^{-tA}B''(\mathrm{e}^{tA}u(t))(A\mathrm{e}^{tA}u(t), B(\mathrm{e}^{tA}u(t)))\\ &\quad + \mathrm{e}^{-tA}B''(\mathrm{e}^{tA}u(t))(B(\mathrm{e}^{tA}u(t)), A\mathrm{e}^{tA}u(t))\\ &\quad + \mathrm{e}^{-tA}B''(\mathrm{e}^{tA}u(t))(B(\mathrm{e}^{tA}u(t)), B(\mathrm{e}^{tA}u(t)).\end{aligned}$$

From these calculations, it is clear that the leading term (associated with the most stringent regularity requirement) involves $\mathrm{e}^{-tA}\mathrm{ad}_A^j(B)(\mathrm{e}^{tA}u(t))$ also for higher derivatives $\frac{\partial^j}{\partial t^j}F(u(t))$, $j \geq 3$. Using the estimates in Lemmas 4.2 and 4.3, we conclude that

$$\begin{aligned}&\|\mathrm{e}^{-tA}[A,B](\mathrm{e}^{tA}u)\|_{H^1} \leq C(\|u\|_{H^2}),\\ &\|\mathrm{e}^{-tA}[A,[A,B]](\mathrm{e}^{t\,A}u)\|_{H^1} \leq C(\|u\|_{H^3}),\\ &\|\mathrm{e}^{-tA}\mathrm{ad}_A^j(B)(\mathrm{e}^{tA}u)\|_{H^1} \leq C(\|u\|_{H^{2j-2}}), \quad j \geq 3.\end{aligned}$$

(3) *Convergence and boundedness in* H^1. From stability and consistency we now conclude convergence in H^1 of order q if $u \in H^2$ for $q = 1$, $u \in H^3$ for $q = 2$ and $u \in H^{2q-2}$ for $q \geq 3$. This also implies that the numerical solution is bounded in H^1.

(4) *Lipschitz condition in* L^2. Under the prerequisite that the numerical solution is bounded in H^1, we can prove an error bound in L^2. From Lemma 4.4, clearly the right-hand side satisfies a Lipschitz condition if the numerical solution is in H^1. This furthermore necessitates that the exact solution be in H^2.

(5) *Local error bound in* L^2. The local error in L^2 can be estimated similarly to the previous considerations. We may assume that the order of the method is greater than one, otherwise no convergence in H^1 could be observed under the same regularity assumptions, the situation is thus comparable to [30, 32]. We can therefore use the estimates

$$\begin{aligned}&\|\mathrm{e}^{-tA}\mathrm{ad}_A^2(B)(\mathrm{e}^{tA}u)\|_{L^2} \leq C(\|u\|_{H^2}),\\ &\|\mathrm{e}^{-tA}\mathrm{ad}_A^j(B)(\mathrm{e}^{tA}u)\|_{L^2} \leq C(\|u\|_{H^{2j-3}}), \quad j \geq 3,\end{aligned}$$

see Lemma 4.3.

(6) *Convergence in* L^2. Stability together with consistency finally yields convergence of order q in L^2 if the exact solution satisfies $u \in H^2$ for $q = 2$ and $u \in H^{2q-3}$, $q \geq 3$.

Altogether, we have thus proven the following convergence result.

Theorem 5.1. *Let the solution u defined by the evolution equations (3.5) be sufficiently regular. Then the numerical solution u_n, $n = 0, 1, \ldots$ defined by (2.4) satisfies the following convergence estimates:*

$$\text{If } u \in H^2, \text{ then } \quad \|u(t_n) - u_n\|_{H^1} = O(h), \qquad p = 1. \tag{5.1}$$

$$\text{If } u \in H^3, \text{ then } \quad \|u(t_n) - u_n\|_{H^1} = O(h^2), \qquad p = 2. \tag{5.2}$$

$$\text{If } u \in H^2, \text{ then } \quad \|u(t_n) - u_n\|_{L^2} = O(h^2), \qquad p = 2. \tag{5.3}$$

$$\text{If } u \in H^{2p-2}, \text{ then } \quad \|u(t_n) - u_n\|_{H^1} = O(h^p), \qquad p \geq 3. \tag{5.4}$$

$$\text{If } u \in H^{2p-3}, \text{ then } \quad \|u(t_n) - u_n\|_{L^2} = O(h^p), \quad p \geq 3. \tag{5.5}$$

6. Numerical illustration

As a numerical illustration, we show the results of an adaptive time propagation of a 1D model of helium in an irradiated laser field as given in [47], where

$$H(t)\Psi(t) = H_0\Psi(t) + (x_1 + x_2)\mathcal{E}(t)\Psi(t), \tag{6.1}$$

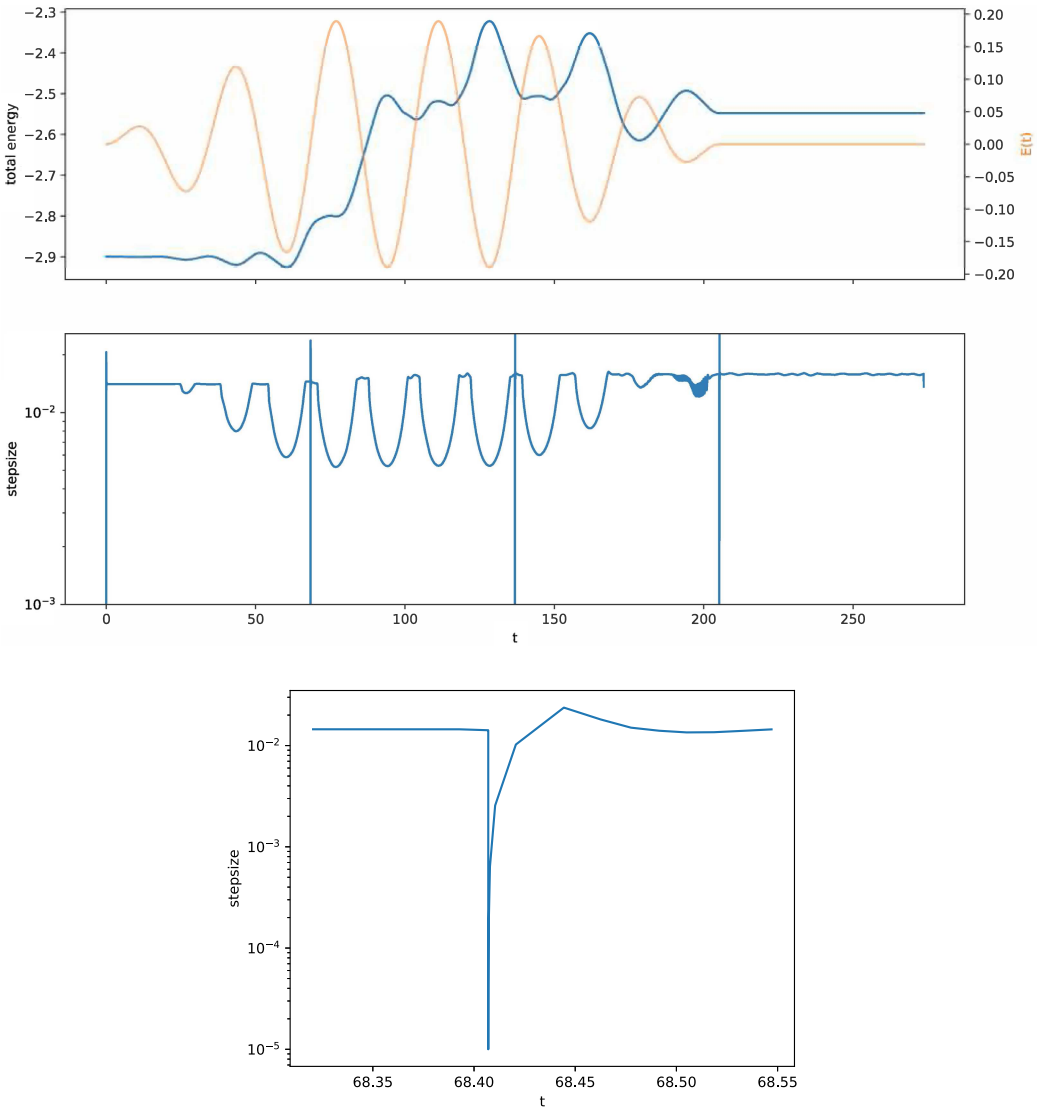

FIGURE 1. Total energy functional and external potential (*top panel*) and automatically generated step-sizes (*middle panel*). Detail near break point (*bottom panel*).

with

$$H_0 = -\frac{1}{2}(\partial_{x_1}^2 + \partial_{x_2}^2) - \frac{2}{\sqrt{x_1^2 + b^2}} - \frac{2}{\sqrt{x_2^2 + b^2}} + \frac{1}{\sqrt{(x_1 - x_2)^2 + b^2}} \tag{6.2}$$

with a smoothed Coulomb potential with shielding parameter $b = 0.7408$, and

$$\mathcal{E}(t) = \mathcal{E}_0 f(t) \sin(\omega t).$$

The peak amplitude is set to $\mathcal{E}_0 = 0.1894$, and frequency $\omega = 0.1837$. We compute the trapezoidal envelope $f(t)$ with 2-cycle turn-on, 2-cycle flat top, and 2-cycle turn-off (cycle duration of laser $= 2\pi/\omega \approx 34.20$), the initial data is given by the ground state of the stationary problem. We prescribe a tolerance of 10^{-5} for the

local error control and show the step-sizes automatically generated by the standard strategy [22]. The spatial discretization is based on Fourier modes associated with 16384 equidistant grid points in the interval $[-512, 512]$. The problem is solved by an adaptive Adams–Lawson multistep propagator of sixth order with local error estimation based on a predictor-corrector strategy. It should be pointed out that this strategy implies only one additional evaluation of the computationally expensive operator B per time step, for a numerical method of any order, and thus provides local error control at minimal cost. Figure 1 shows the external potential $E(t)$ and total energy functional (top) illustrating the local solution smoothness, and the step-sizes (middle). The unsmooth envelope of the time-dependent potential enforces two restarts which implies artificially small step-sizes and a subsequent overshoot until the adaptive strategy chooses consistent step-sizes again. This phenomenon is shown at higher resolution in the bottom plot. The automatically chosen step-sizes reflect the solution's smoothness, being smallest at the maxima and minima of the functional and returning to (almost) constant steps after the external potential subsides. This behavior supports the correctness of the theoretical convergence results which are the subject of the present paper.

7. Conclusions

We have analyzed the convergence of exponential Lawson multistep methods for the time integration of the equations of motion associated with the multiconfiguration time-dependent Hartree–Fock approximation for high-dimensional electron dynamics. The methods show their classical convergence order under moderate assumptions on the solution regularity. The consistent behavior of the methods has been verified experimentally in [3] and is illustrated in the present work, which shows the success of an adaptive predictor-corrector method, indicating that the theoretical orders pertain in practical simulations.

Acknowledgements. The author would like to acknowledge support with the numerical illustrations by Harald Hofstätter (Universität Wien) and Alexander Grosz (student at TU Wien). This work was supported by the Vienna Science and Technology Fund (WWTF) under the grant MA14-002.

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