EFFICIENT COMPUTATION OF THE MCTDHF APPROXIMATION TO THE TIME-DEPENDENT SCHRÖDINGER EQUATION

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Abstract. We discuss analytical and numerical properties of the multi-configuration time-dependent Hartree–Fock method for the approximate solution of the time-dependent multi-particle (electronic) Schrödinger equation which are relevant for an efficient implementation of this model reduction technique. Particularly, we focus on a discretization and low rank approximation in the evaluation of the meanfield terms occurring in the MCTDHF equations of motion, which is crucial for the computational tractability of the problem. We give error bounds for this approximation and demonstrate the achieved gain in performance.

1. Introduction

In this paper we discuss analytical and numerical aspects of the multi-configuration time-dependent Hartree–Fock method (MCTDHF) for the approximate solution of the time-dependent Schrödinger equation

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

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

$$H(t) := \sum_{k=1}^{f} \left(\frac{1}{2} \left(-\mathrm{i}\nabla^{(k)} - A(t) \right)^2 + U(x^{(k)}) + \sum_{l<k} V(x^{(k)} - x^{(l)}) \right), \tag{2}$$

where

$$U(x) := -\frac{Z}{|x|}, \quad Z \in \mathbb{N}, \tag{3}$$

$$V(x-y) := \frac{1}{|x-y|}, \tag{4}$$

$$A(t) := (a_1(t), a_2(t), a_3(t)). \tag{5}$$

$A(t)$ is a smooth (vector-valued) function of t modeling an ultrafast laser pulse, and $\nabla^{(k)}$ is the nabla operator w. r. t. $x^{(k)}$ only.

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1.1. **The MCTDHF Method.** In MCTDHF as put forward in [6, 27, 28], the multi-electron wave function ψ from (1) is approximated by a function satisfying the ansatz

$$u = \sum_{(j_1,\dots,j_f)} a_{j_1,\dots,j_f}(t)\phi_{j_1}(x^{(1)},t)\cdots\phi_{j_f}(x^{(f)},t) =: \sum_J a_J(t)\Phi_J(x,t). \tag{6}$$

Using (6) 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 [2, 3, 20, 21] 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 [14] that this does not change our considerations concerning the equations of motion associated with MCTDHF. Consequently, we ignore spin and concentrate on the representation (6).

The Dirac–Frenkel variational principle [8, 10] is used to derive differential equations for the coefficients a_J and the *single-particle functions* ϕ_j. Thus, for u in

$$\mathcal{M} = \left\{ u : u(x,t) = \sum_{(j_1,\dots,j_f)} a_{j_1,\dots,j_f}(t)\phi_{j_1}(x^{(1)},t)\cdots\phi_{j_f}(x^{(f)},t) \right\}, \tag{7}$$

where $a_J(t) \in \mathbb{C}$ and $\phi_{j_k}(\cdot,t) \in L^2$, we require

$$\left\langle \delta u \left| \mathrm{i}\frac{\partial}{\partial t} - H \right| u \right\rangle_f = 0, \tag{8}$$

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

$$\mathcal{T}_u\mathcal{M} = \left\{ \delta u : \delta u(x,t) = \sum_J \left(\delta a_J \Phi_J(x,t) + a_J(t) \sum_{k=1}^{f} \delta\phi_{j_k}(x^{(k)}) \prod_{l\neq k} \phi_{j_l}(x^{(l)},t) \right) \right\},$$

with $\delta a_J \in \mathbb{C}$ and $\delta\phi_{j_k} \in L^2$. By $\langle\cdot|\cdot\rangle_f$ we denote the usual inner product in L^2 w. r. t. f spatial variables. Recall that for an operator H,

$$\langle\psi|H|\tilde\psi\rangle_f = \langle\psi|H\tilde\psi\rangle_f = \int\cdots\int \overline{\psi}(H\tilde\psi)\,dx^{(1)}\cdots dx^{(f)}.$$

We will subsequently also use inner products with subscripts 1, 2 or $f-1$ to denote inner products over the respective number of degrees of freedom.

It was shown in [16] that the set $\mathcal{M}$ in conjunction with a full-rank-condition for the *density matrix* ρ defined in (14) 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 sloppily refer to $\mathcal{M}$ as a manifold under the assumption that ρ is nonsingular.

In order to define a unique solution of (8), we impose additional constraints,

$$\left\langle \phi_j \middle| \phi_k \right\rangle_1 = \delta_{j,k}, \quad t \geq 0, \tag{9}$$

$$\left\langle \phi_j \middle| \frac{\partial \phi_k}{\partial t} \right\rangle_1 = 0, \quad t \geq 0. \tag{10}$$

The variational principle (8) and the additional restrictions (9), (10) finally yield equations of motion for the coefficients and single-particle functions in (6):

$$\mathrm{i}\frac{da_J}{dt} = \sum_K \left\langle \Phi_J \left| H \right| \Phi_K \right\rangle_f a_K, \quad \forall J, \tag{11}$$

$$\mathrm{i}\frac{\partial \phi_j}{\partial t} = (I - P) \sum_{k=1}^{N} \sum_{l=1}^{N} \rho_{j,l}^{-1} \overline{H}_{l,k} \phi_k, \quad j = 1, \ldots, N, \tag{12}$$

where

$$\psi_j := \left\langle \phi_j | u \right\rangle_1, \tag{13}$$

$$\rho_{j,l} := \left\langle \psi_j | \psi_l \right\rangle_{f-1}, \tag{14}$$

$$\overline{H}_{j,l} := \left\langle \psi_j \left| H \right| \psi_l \right\rangle_{f-1}, \tag{15}$$

and P is the orthogonal projector onto the space spanned by the functions ϕ_j.

For the remainder of this paper, we will focus on the solution of the system (11), (12). After stating analytical prerequisites like existence, uniqueness and regularity of the solutions of (1) and (11), (12) in §1.2, we will briefly discuss the numerical methods used for the equations of motion in §1.3. Section 2 is devoted to a technical detail which is crucial for the successful numerical solution of (11), (12): in the right-hand side of (12), the evaluation of the *meanfield operator matrix* $\overline{H}$ according to (15) constitutes the computationally most expensive part of the algorithm. A discretization and low rank approximation in the evaluation of the involved integrals which was proposed in [6] serves to make the problem computationally tractable. In §2 we analyze this approximation, and demonstrate that at the cost of a moderate loss of precision, a big gain in efficiency may be achieved. The main new result of this paper is that the approximation is justified theoretically and error bounds can be given, thus ensuring a reliable evaluation of the differential equations (11), (12).

1.2. **Analytical Results.** Before turning to the numerical treatment of the MCTDHF equations of motion, we discuss the question of the well-posedness of the problem at hand (existence, uniqueness and regularity of the solution, as well as smooth dependence on the initial data, cf. [9, Def. VI.9.1]). This is an important prerequisite for the analysis of numerical methods. Both in the study of the integration of (11), (12) according to §1.3, and the analysis of the approximation described in §2, the (spatial) regularity of the single-particle functions ϕ_j is an essential requirement. More precisely, we demand that the solutions are in certain *Sobolev spaces* H^m for $m \geq 1$. The Sobolev space H^1 consists of all $\psi \in L^2$ with

$$\|\psi\|_{H^1} := \|\psi\| + \||\nabla \psi|\| < \infty.$$

We will also consider the space $H^2 \subseteq L^2$, where we require

$$\|\psi\|_{H^2} := \|\psi\| + \||\nabla \psi|\| + \|\Delta \psi\| < \infty,$$

and similarly for H^m with $m > 2$. All derivatives are meant in the *weak sense*, see for example [5, Ch. 1]. $\|\cdot\|$ denotes the usual norm in L^2.

First, to demonstrate the well-posedness of (1) we use [9, VI.9.5]. To this end, [24, Cor. X.4.2] is used to show that $H(t)$ is essentially self-adjoint for all t. For $A(t) \equiv 0$, denote by D the domain of the closure of H. What remains to be shown is that the closure of $H(t)$ has the same domain for every t. This is proven with [23, Thm. X.12] on noting that ∇ is infinitesimally small with respect to Δ [23, Thm. X.22]. From results in [11] and [22] it is clear that $D = H^2$. Now, if $A(t) \in C^1$, then $\dot{A}(t)$ in place of $A(t)$ in (2) defines a perturbation which can be treated analogously as above. Thus we conclude that the corresponding operator is self-adjoint and maps H^2 into L^2. Consequently, $\dot{H}(t)$ is well-defined and $t \mapsto H(t)x$ is differentiable w. r. t. t for every $x \in H^2$, and the requirements of [9, VI.9.5] are satisfied. Consequently, there exists a unique classical solution $\psi \in H^2$ of (1) for all $t \geq 0$ which depends smoothly on the initial data.

Next, we turn to the well-posedness of the equations of motion (11), (12). This point is crucial for the numerical treatment of the MCTDHF equations. If the potential part $U + V$ in (2) is smooth, a regularity result from [16] carries over to the case of MCTDHF. This can be stated as follows:

Theorem 1.1. *Consider the system* (11), (12) *together with initial conditions chosen such that the orthonormality constraints* (9) *are satisfied and the density matrix* ρ *defined in* (14) *is nonsingular. Assume that the potential* $U + V$ *in* (2) *is bounded and twice continuously differentiable, with bounded first and second derivatives. If the initial data for* ϕ_j *is in the Sobolev space* H^2*, then there is a* $t^* > 0$ *such that for* $t \in [0, t^*]$ *there exists a unique classical solution of the MCTDHF equations* (11), (12) *satisfying*

$$a_J \in C^2([0,t^*],\mathbb{C}), \quad \phi_j \in C^1([0,t^*],L^2) \cap C([0,t^*],H^2).$$

The solution can be continued in time until the density matrix ρ *becomes singular. It depends Lipschitz continuously with respect to the* H^2 *norm on the initial data. Moreover, for* u *defined by* a_J, ϕ_j *via* (6)*, we have* $u(t) \in H^2$ *for* $t \in [0,t^*]$*, and* u *solves the Dirac–Frenkel variational equation* (8)*.*

It is important to note that the above theorem cannot guarantee the existence of the solution u for all times $t > 0$. Rather, the equations (11), (12) break down when the density matrix ρ becomes singular. It is the subject of ongoing investigation whether this may actually happen, and under what circumstances such a breakdown is possible. In practice, a regularization is employed to prevent the algorithm from failing, see [2, 16]. We would like to stress however that the assumption on ρ is crucial for the whole approach, as the set $\mathcal{M}$ from (7) does not represent a manifold if ρ becomes singular.

Theorem 1.1 can easily be extended to yield higher regularity of the solution, $u \in H^m$ for $m > 2$, if the initial data has the same regularity and the potential is sufficiently smooth with bounded derivatives [16]. However, for the unbounded potential in (2), such a result has not been proven yet. An extension of Theorem 1.1 to this case is a major focus of ongoing research.

In practical model computations, regularized potentials are often employed. For instance, [6] treats examples in one spatial dimension with a *screened Coulomb potential*, see also [14]. In these situations, the regularity results from [16] carry over. This has important consequences for the approximation properties of the MCTDHF approach. According to [19, Theorem 4.1], the error of the MCTDHF approximation to the exact wave function is — for sufficiently short time intervals — of the same order of magnitude as the error of the best approximation in the

approximation manifold. This result requires H^2 regularity of the approximate wave function. The regularity is also needed to ensure good performance of numerical methods used to solve the MCTDHF equations, both for space discretization and for the variational splitting integrator of [18] for the discretization in time. In §1.3, we will give a short description of numerical methods for the integration of (11), (12) and explain the role of regularity in more detail.

1.3. **Numerical Treatment.** To compute the solution of (11), (12) numerically, the method of lines is used. First, space discretization is applied to derive a system of ordinary differential equations. In our code [6] which is designed for problems in one space dimension, we use the *pseudospectral method* [26] at a uniform grid on a suitably truncated domain. We found that this method is advantageous as compared with space discretization by finite differences. Firstly, for problems with a sufficiently regular solution, the error decreases exponentially when the spatial grid is refined [26]. Moreover, the spectrum of the differential operator is approximated well even by the eigenvalues of its discretization with large modulus [17]. For numerical comparisons, see [12, 13]. To achieve a good rate of convergence of the approximation, a highly regular solution u is desirable. Theorem 1.1 and its extension to higher regularity provides the necessary theoretical background.

For spatial dimension higher than one, parallelization becomes an important issue. In that case, the pseudospectral method is not optimal due to the non-locality of the basis functions, and finite elements appear favorable for space discretization [7]. In this case, an adaptive choice of non-uniform spatial grids should be considered (this does not make sense in general when the pseudospectral method is used). The extension of the existing MCTDHF code to higher dimension is currently work in progress.

By default, we use explicit Runge-Kutta methods for the time integration of the ODEs resulting after space discretization. For reasonably smooth data and moderate spatial grid spacing, these methods were found to work dependably and retain their classical convergence orders, thereby yielding an efficient method for time integration. Extensive test results reported in [13] support this claim, see also [12].

For difficult problems and fine spatial grids, however, a more robust, low-order alternative is given by *variational splitting* introduced in [18] for the nonlinear PDEs arising from MCTDH in quantum molecular dynamics. This method is based on a symmetric operator splitting of the Hamiltonian $H(t)$ from (2), commonly referred to as *Strang splitting*. This makes it possible to treat separately the *kinetic part* consisting of the unbounded differential operator and the potential part $U + V$. In many situations, this can be exploited to reduce the number of evaluations of the meanfield operators by choosing larger time steps in the integration of the potential part. This is a crucial factor for the tractability of the computational problem, see §2. Variational splitting yields a second order approximation for the solution of the full problem (8) if the potential is smooth with bounded derivatives and if $u \in H^2$, see [18]. Thus the regularity result in Theorem 1.1 is an important prerequisite to derive error bounds for this time integrator.

2. Approximation of the Meanfield Operators

To make the numerical solution of the equations of motion (11), (12) computationally tractable, some care is required in the evaluation of the right-hand side of the

differential equations. The computationally most demanding part is represented by the evaluation of the meanfield operators (15), see also [15, Fig. 2.3]. This becomes clear when we realize that the computations involve the evaluation of integrals of the form

$$\langle\phi_1|V|\phi_2\rangle_2 = \langle\phi_1(r)|V(r-r')|\phi_2(r')\rangle_2 = \int_{\mathbb{R}^3}\int_{\mathbb{R}^3} \overline{\phi_1(r)}V(r-r')\phi_2(r')\,dr'\,dr, \tag{16}$$

where $\phi_1,\ \phi_2$ are any single-particle functions from (6). Note that it is necessary to compute a considerable number of evaluations of integrals (16): the contributions from a large set of single-particle functions are necessary because the system under consideration is highly correlated [6].

To reduce the computational effort necessary for the evaluation of (16), a procedure based on discretization and low rank approximation is proposed in [6], which approximates the full integral in $\mathbb{R}^6$ as a sum (of moderate length) of products of integrals in $\mathbb{R}^3$. Here, we are going to derive bounds for the error introduced by this approximation.

Following [6], we choose a discretization in terms of a set of basis functions $\mathcal{B} = \{|i\rangle = i(r) : i = 1,\dots,L\}$ and approximate V by

$$V \approx V_{\text{app}} = RVR, \tag{17}$$

where R is the orthogonal projection onto the subspace B spanned by $\mathcal{B}$. As basis functions we use real-valued functions with finite support defined on a subgrid of the spatial grid used to solve (11), (12). In general, it is natural to use polynomial finite elements on a suitable subdivision of the spatial domain. The details may vary from case to case. Here, we attempt to give a general idea of what error bounds can be expected if the space partition is sensible and the choice of finite elements corresponds with the task at hand. The notions used in the subsequent presentation are explained for example in [5].

We consider for $\mathcal{B}$ the *nodal basis* for globally continuous finite elements consisting of piecewise polynomials of degree $\leq m-1$. The resulting finite elements are *conforming*, i. e., $B \subseteq H^1$ [4, Thm. II.5.2]. For simplicity and ease of presentation, we will henceforth mostly address the special case of finite elements with $m = 2$ in one spatial dimension. These were also used in the computations reported in [6]. However, our results extend easily to the more general case laid out above. We will state the corresponding error bounds at appropriate places and refer to the auxiliary results we require from the literature for the general case. Note that in an extension of the code [6] which is currently being developed for problems in two spatial dimensions, tensor products of the one-dimensional finite elements defined in [25] are used. Our results trivially carry over in that case as well, since the approximation errors are bounded from below by the best approximations in two dimensions and from above in terms of the bounds for one dimension. Since these have the same asymptotic behavior as the diameter of the space partition goes to zero, the tensor products yield analogous results and we will not discuss this special case further.

In the case of one space dimension, we consider an underlying uniform spatial grid $\Delta = (x_{-K},\dots,x_K)$ on a suitably truncated interval $[-x_{\text{end}}, x_{\text{end}}]$. We will not discuss the (negligible) error introduced by the truncation in this paper. We heuristically choose a subgrid $\Gamma = (\tau_0 = x_{-K},\dots,\tau_{L+1} = x_K)$ of Δ, where τ_j are denser near the core where solutions vary more rapidly (our analysis below shows that interval lengths should be smaller where $\nabla^2\phi_i,\ i = 1,2$ is large). Now, the

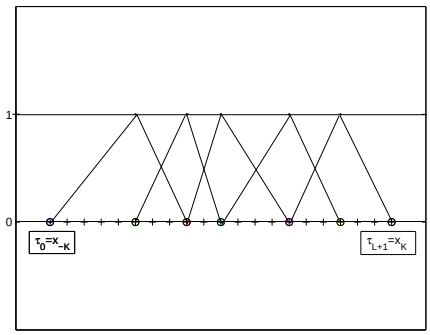

FIGURE 1. The hat functions representing the nodal basis for linear finite elements in one spatial dimension.

basis function $|i\rangle$ is the continuous, piecewise linear function with $i(\tau_j) = \delta_{j,i}$. This is the *nodal basis* for the piecewise linear, continuous finite element approximation on Γ. Note that trivially, $|i\rangle \in H^1$ holds. The *hat functions* $|i\rangle$ are illustrated in Figure 1. We will always assume that our spatial subdivision is *quasi-uniform* [5, Def. (4.4.13)], which for the grid Γ means that for some (moderate) constants $\tilde{\kappa}, \kappa$,

$$\tilde{\kappa}h \leq \tau_{j+1} - \tau_j \leq \kappa h, \quad h := \frac{1}{L}, \tag{18}$$

holds for all L.

Now, the projection R used in (17) is characterized by

$$R = \sum_{i,j=1}^{L} |i\rangle [Q^{-1}]_{i,j} \langle j|, \tag{19}$$

with the *mass matrix* Q given as

$$Q_{i,j} = \langle i|j\rangle_1. \tag{20}$$

Obviously, Q is real, symmetric and nonsingular, and thus Q^{-1} is also symmetric. It is easy to show that $R^2 = R$ and R is symmetric, $\langle \phi_1 | R\phi_2\rangle_1 = \langle R\phi_1|\phi_2\rangle_1$. Consequently, R is indeed the orthogonal projection onto B. Note also that, since R is bounded, R is self-adjoint.

Since the (real) multiplication operator V is self-adjoint, the approximation error resulting from our discretization can be estimated as

$$\begin{aligned} &|\langle \phi_1 | V - V_{\mathrm{app}} | \phi_2 \rangle_2| \\ &\quad = |\langle \phi_1 | V | \phi_2 - R\phi_2 \rangle_2 + \langle \phi_1 - R\phi_1 | V | R\phi_2 \rangle_2| \\ &\quad = |\langle V\phi_1 | \phi_2 - R\phi_2 \rangle_2 + \langle \phi_1 - R\phi_1 | VR\phi_2 \rangle_2| \\ &\quad \leq \begin{cases} \text{const.}\,(\|\phi_1\|\|\phi_2 - R\phi_2\| + \|\phi_1 - R\phi_1\|\|R\phi_2\|) & \text{for } V \text{ bounded,} \\ \text{const.}\,(\|\phi_1\|_{H^1}\|\phi_2 - R\phi_2\| + \|\phi_1 - R\phi_1\|\|R\phi_2\|_{H^1}) & \text{for } V \text{ Coulomb.} \end{cases} \end{aligned} \tag{21}$$

For the estimate (21), see for example [19]. Note that of course the bound for the Coulomb potential is particular to spatial dimension three, while the estimate for smooth potentials is the same in any dimension.

For our further analysis we assume that $\phi_i \in H^2,\ i = 1, 2$. Sufficient conditions for the regularity of the single-particle functions in the case of bounded potentials (as is the case in our one-dimensional example, see [6]) have been given in Theorem 1.1.

We will now discuss the error committed by projecting a function $\phi \in H^2$ (truncated to the domain of integration) to B for the case of one spatial dimension. In that case, $\phi(x) = 0$ for $|x| \geq x_{\text{end}}$.

The space B consists of all continuous, piecewise linear functions on Γ which vanish for $|x| \geq x_{\text{end}}$. Since R is the orthogonal projection onto this space, $R\phi$ represents the best approximation in B w. r. t. the norm in L^2. Consequently,

$$\begin{aligned} \int_{-x_{\text{end}}}^{x_{\text{end}}} |\phi(x) - R\phi(x)|^2 \, dx &= \sum_{j=0}^{L} \int_{\tau_j}^{\tau_{j+1}} |\phi(x) - R\phi(x)|^2 \, dx \\ &\leq \sum_{j=0}^{L} \int_{\tau_j}^{\tau_{j+1}} |\phi(x) - P\phi(x)|^2 \, dx, \end{aligned} \tag{22}$$

where $P\phi$ is the continuous, piecewise linear interpolant of ϕ at Γ. According to [5, Cor. (1.4.7)], every $\phi \in H^2$ can be represented by a continuous function, and thus the interpolation is well defined. According to [5, Cor. (4.4.24)],

$$\int_{\tau_j}^{\tau_{j+1}} |\phi(x) - P\phi(x)|^2 \, dx \leq \text{const.} \, (\tau_{j+1} - \tau_j)^4 \int_{\tau_j}^{\tau_{j+1}} |\nabla^2 \phi(x)|^2 \, dx. \tag{23}$$

Substituting (23) into (22), we obtain

$$\|\phi - R\phi\|^2 \leq \text{const.} \sum_{j=0}^{L} \|\nabla^2 \phi\|^2_{[\tau_j, \tau_{j+1}]} (\tau_{j+1} - \tau_j)^4, \tag{24}$$

where $\| \cdot \|_{[\tau_j, \tau_{j+1}]}$ denotes the L^2 norm on the interval $[\tau_j, \tau_{j+1}]$. Thus, the conclusions from this error analysis are:

(1) The intervals $[\tau_j, \tau_{j+1}]$ should be chosen smaller where $\nabla^2 \phi$ is large.
(2) We can conclude an overall bound

$$\|\phi - R\phi\| = O(h^2),$$

see also [5, Remark (4.4.27)]. Thus, for a bounded potential, an error bound of $O(h^2)$ results in (21).

To complete the estimate of the discretization error for the Coulomb potential, we have to consider $\|R\phi\|_{H^1}$. From [5, Thm. (4.5.11)] and the remark following the proof of this proposition, it is clear that $\|R\phi\|_{H^1} = O(1/h)$.

[5, Cor. (4.4.24)] indicates that the convergence order increases if more regularity on ϕ is assumed and the basis functions are chosen as polynomials of higher degree $m - 1 > 1$. Namely, $\|\phi - R\phi\| \leq \text{const.} \, h^m \|\nabla^m \phi\|$ holds for $\phi \in H^m$. This result holds generally also in spatial dimensions greater than one. We sum up the results of our previous analysis and this last observation in the following theorem.

Theorem 2.1. *Let $\mathcal{B}$ be the nodal basis for globally continuous, piecewise polynomial finite elements of degree $m - 1$ on a suitable, quasi-uniform spatial partition with diameter equal to h. Then, for the discretization error committed in the approximation* (17)*, the bounds*

$$|\langle \phi_1 | V - V_{\text{app}} | \phi_2 \rangle_2| = \begin{cases} O(h^m) & \text{for } V \text{ bounded,} \\ O(h^{m-1}) & \text{for } V \text{ Coulomb} \end{cases}$$

hold.

As the last step in our error analysis, we consider the low rank approximation of the discrete operator V_{app} according to [6].

First, we rewrite (17) as

$$V_{\text{app}} = \sum_{i,j=1}^{L} \sum_{i',j'=1}^{L} |i\rangle [Q^{-1}]_{i,i'} \tilde{V}_{i',j'} [Q^{-1}]_{j',j} \langle j| = \sum_{i,j=1}^{L} |i\rangle [Q^{-1} \tilde{V} Q^{-1}]_{i,j} \langle j|$$

with

$$\tilde{V}_{i,j} = \int \int i(r) V(r - r') j(r') \, dr' \, dr.$$

Obviously, the matrix $\tilde{V}$ is symmetric. We now rewrite $\tilde{V}$ as follows: define a matrix S by

$$S_{i,j} := \int i(r) g(r) j(r) \, dr$$

with a real function $g > 0$ which is large in regions where we wish to put an emphasis in our computations, usually near the core. Obviously, S is symmetric, and it is easy to see that S is positive definite. We now consider the generalized eigenvalue problem

$$\tilde{V} u = \lambda S u.$$

We can rewrite this problem as an ordinary eigenvalue problem by using the properties of S. Since S is symmetric positive definite, we can compute the Cholesky factorization

$$S = \tilde{C}^T \tilde{C},$$

with a nonsingular upper triangular matrix $\tilde{C}$. Setting $\tilde{u} := \tilde{C} u$ we obtain

$$(\tilde{C}^T)^{-1} \tilde{V} \tilde{C}^{-1} \tilde{u} = \lambda \tilde{u}.$$

This eigenvalue problem has a symmetric matrix, and thus an orthogonal basis $\tilde{U}$ of eigenvectors exists to the real eigenvalues $\lambda_1, \ldots, \lambda_L$. It is straightforward to show that

$$\tilde{V} = \hat{U} \Lambda \hat{U}^T,$$

where

$$\hat{U} := \tilde{C}^T \tilde{U}, \quad \Lambda = \text{diag}(\lambda_1, \ldots, \lambda_L).$$

Now we discard the contributions associated with eigenvalues with small modulus. For simplicity of notation assume that $\lambda_1 \geq \lambda_2 \geq \cdots \geq \lambda_L > 0$, $\lambda_i < \varepsilon$ for $i > M$, where ε is an error margin which can be freely chosen. Setting

$$\Lambda_\varepsilon := \text{diag}(\lambda_1, \ldots, \lambda_M, 0, \ldots, 0), \quad \tilde{V}_\varepsilon := \hat{U} \Lambda_\varepsilon \hat{U}^T,$$

we approximate V_{app} by

$$\begin{aligned} V_{\text{app}} \approx V_{\text{low}} &:= \sum_{i,j=1}^{L} |i\rangle [Q^{-1} \tilde{V}_\varepsilon Q^{-1}]_{i,j} \langle j| \\ &= \sum_{\mu=1}^{M} \sum_{i,j=1}^{L} |i\rangle [Q^{-1} \hat{U}]_{i,\mu} \lambda_\mu [\hat{U}^T Q^{-1}]_{\mu,j} \langle j|. \end{aligned} \tag{25}$$

Thus we have achieved our goal to find a representation of V in terms of a reasonably short sum of products of potentials that depend on only one spatial variable. The

error introduced by the low rank approximation can be estimated as

$$\begin{aligned}
|\langle\phi_1|V_{\text{app}} - V_{\text{low}}|\phi_2\rangle_2| &= \left|\sum_{i,j=1}^{L} \langle\phi_1|i\rangle_1\langle j|\phi_2\rangle_1 \sum_{\mu=M+1}^{L} [Q^{-1}\hat{U}]_{i,\mu}\lambda_\mu[\hat{U}^T Q^{-1}]_{\mu,j}\right| \\
&\leq \|\phi_1\|\|\phi_2\| \sum_{i,j=1}^{L} \|i\|\|j\| \left|\sum_{\mu=M+1}^{L} [Q^{-1}\hat{U}]_{i,\mu}\lambda_\mu[\hat{U}^T Q^{-1}]_{\mu,j}\right| \\
&\leq \text{const.}\, \varepsilon\, h^2 \sum_{i,j=1}^{L}\sum_{\mu=M+1}^{L} |[Q^{-1}\hat{U}]_{i,\mu}||[\hat{U}^T Q^{-1}]_{\mu,j}| \\
&\leq \text{const.}\, \varepsilon h^2 \sum_{i,j=1}^{L} [|Q^{-1}\hat{U}||\hat{U}^T Q^{-1}|]_{i,j} \\
&\leq \text{const.}\, \varepsilon h^2 L \|Q^{-1}\hat{U}\|_2^2 \leq \text{const.}\, \varepsilon h^2 L \|Q^{-1}\|_2^2 \|\hat{U}\|_2^2 \\
&\leq \text{const.}\, \varepsilon,
\end{aligned}$$

where the absolute value of a matrix, $|A|$, is meant entry-wise. In the estimates above we have used the following properties: since the spatial partition is quasi-uniform, $Lh \leq$ const. Trivially, $\|i\| = O(h)$ for the basis functions $|i\rangle$. For the estimation of the involved matrices, we use the *spectral norm* defined for $A \in \mathbb{R}^{L\times L}$ as

$$\|A\|_2 = \max_{i=1,\ldots,L} \sigma_i,$$

where σ_i are the *singular values* of A. From [1, Sec. 5.5] we have $\|Q^{-1}\|_2 = O(1/h)$, and moreover

$$\|\hat{U}\|_2^2 = \|S\|_2 = O(h).$$

The last assertion follows from

$$\hat{U}^T\hat{U} = \tilde{U}^T\tilde{C}\tilde{C}^T\tilde{U} = \tilde{U}^T\tilde{C}S\tilde{C}^{-1}\tilde{U},$$

whence $\hat{U}^T\hat{U}$ and S have the same eigenvalues, which for symmetric positive definite matrices correspond with the singular values. Note that $\|S\|_2 = O(h)$ follows from [1, Sec. 5.5] because S is the mass matrix associated with the basis functions $\sqrt{g}|i\rangle$. Finally, we have used a simple property that is formulated in the next lemma:

Lemma 2.1. *Let $X \in \mathbb{R}^{L\times L}$ be a nonsingular matrix. Then,*

$$\sum_{i,j=1}^{L} [|X^T||X|]_{i,j} \leq L\|X\|_2^2.$$

Proof. Under the assumptions of the lemma, X^TX is symmetric positive definite, and consequently

$$\|X\|_2^2 = \|X^TX\|_2 = \max_{0\neq y\in\mathbb{R}^L} \frac{y^TX^TXy}{y^Ty},$$

see for instance [1, Sec. 5.5]. Choose y with $y_k = \pm 1$ such that $[Xy]_k = \sum_{i=1}^{L} |x_{k,i}|$. It is easy to see that

$$\frac{y^TX^TXy}{y^Ty} = \frac{1}{L}\sum_{k=1}^{L}\left(\sum_{i=1}^{L}|x_{k,i}|\right)^2 = \frac{1}{L}\sum_{i,j,k=1}^{L}|x_{k,i}||x_{k,j}| = \frac{1}{L}\sum_{i,j=1}^{L}[|X^T||X|]_{i,j}. \qquad \square$$

Thus, the error of our low rank approximation is bounded in terms of the error margin ε, uniformly in the diameters of the spatial partition. We formulate this fact in the following theorem:

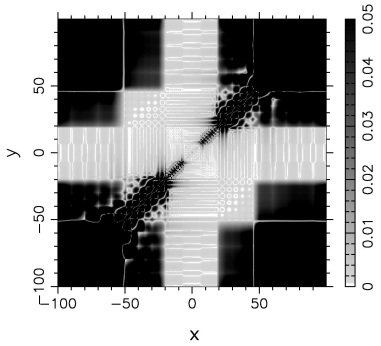

FIGURE 2. Relative error $|V - V_{\text{low}}|/|V|$.

Theorem 2.2. *For $\phi_1,\ \phi_2 \in L^2$, and an underlying spatial partition which is quasi-uniform, the error introduced by the low rank approximation* (25) *satisfies*

$$|\langle\phi_1|V_{\text{app}} - V_{\text{low}}|\phi_2\rangle_2| = O(\varepsilon)$$

independently of the diameter of the spatial partition, where ε is an error margin which can be freely chosen.

To conclude this section, we point out that the approximation of the meanfield operators described above significantly reduces the computational effort needed to solve the MCTDHF equations, while maintaining an acceptable level of accuracy. This is illustrated by an example given in [6]: on an underlying equidistant grid $\Delta = (x_{-K}, \ldots, x_K)$ with $K = 500$ in one spatial dimension, the (heuristically chosen) subgrid $\Gamma = (\tau_0, \ldots, \tau_{L+1})$ was used to define $L = 83$ basis functions, and yielded V_{low} with $M = 55$. Thus, the computational effort was significantly reduced. Still, the error in the region of interest remained acceptably small. Figure 2 shows the relative error introduced by our approximation, where the grayscale indicates the magnitude of $|V(x-y) - V_{\text{low}}(x-y)|/|V(x-y)|$. In the inner cross-shaped region of interest, the relative error is below one percent. The figure is taken from [6], where the example is explained in more detail.

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