A Hybrid EM/QM Framework Based on the ADHIE-FDTD Method for the Modeling of Nanowires

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Abstract—A new modeling formalism to compute the time-dependent behavior of combined electromagnetic (EM) and quantum mechanical (QM) systems is proposed. The method is geared towards highly multiscale geometries, which is vital for the future design of nanoelectronic devices. The advocated multiphysics modeling formalism leverages the alternating-direction hybrid implicit-explicit (ADHIE) finite-difference time-domain (FDTD) method for the EM fields and is combined with a novel ADHIE method for the EM potentials. Additionally, we tackle the QM problem using a new split real and imaginary part formulation that includes higher-order spatial differences and arbitrary time-dependent EM potentials.

The validity of the proposed formalism is theoretically discussed by deriving its stability condition and calculating the numerical dispersion relation. Furthermore, the applicability of our modeling approach is proven through several numerical experiments, including a single-particle Maxwell-Schrödinger (MS) system by deriving its stability condition and calculating the numerical dispersion relation. Furthermore, the applicability of our modeling approach is proven through several numerical experiments, including a single-particle Maxwell-Schrödinger (MS) system as well as a many-particle Maxwell-Kohn-Sham (MKS) system within the time-dependent density-functional theory (TDDFT) framework. These experiments confirm that the novel ADHIE method drastically decreases the computational time while retaining the accuracy, leading to efficient and accurate simulations of light-matter interactions in multiscale nanoelectronic devices.

Index Terms—Alternating-direction implicit (ADI), finite-difference time-domain (FDTD), hybrid implicit-explicit (HIE), Maxwell, Schrödinger, time-dependent density-functional theory (TDDFT).

I. INTRODUCTION

The modeling of the interaction between electromagnetic (EM) waves and matter on a nanoscale level is gaining importance as devices get ever smaller and incorporate novel materials into their design [1]–[4]. To accurately characterize the dynamical behavior of nanoelectronic components, one relies on numerical simulation techniques that can accurately and efficiently solve the pertaining equations.

Up to now, most methods dealt with either the EM problem or the quantum mechanical (QM) problem. The finite-difference time-domain (FDTD) method is one of the most popular methods for computational EM (CEM). However, in multiscale problems, the small spatial cells lead to an oversampling in time, slowing down the computation. Therefore, many implicit FDTD schemes have been proposed that are unconditionally stable [5]–[7]. The main problem is that the implicitization results in more computationally expensive time stepping. Recently, these techniques have been modified to apply the implicitization locally so that specific cells can be eliminated from the stability criterion on nonuniform grids [8]. As a result, the time step is increased while minimizing the computational cost per time step, resulting in a much more efficient scheme.

The FDTD method is also popular in computational QM and has been applied to the Schrödinger or Kohn-Sham equations with many variations [9]–[14]. Implicit schemes such as [15], [16] have been recently improved by also including local implicitization [17].

Many implementations exist that couple the EM and QM problems [18]–[21], where, depending on the problem at hand, an effective-mass Schrödinger equation [22]–[26] or the \textit{ab initio} Kohn-Sham equation [27]–[30] is considered. However, the coupling of QM and EM systems usually results in a large mismatch between the maximum allowed time step of the respective systems because of the multiscale character. So far, no computationally efficient methods that mitigate this mismatch have been proposed.

In this paper, we propose a new scheme for solving Maxwell-Schrödinger or Maxwell-Kohn-Sham systems. It is specifically designed to tackle the multiscale geometries often present in nanoelectronic devices. For the EM part of the calculation it leverages the alternating-direction hybrid implicit-explicit (ADHIE) FDTD method [8] for the electric \textbf{E} and magnetic \textbf{H} fields. In contrast to the regular Yee-method, where small spatial steps would strongly decrease the allowed time step, the ADHIE method removes particular steps from the stability criterion resulting in a much larger overall time step. Furthermore, a novel formalism is developed to compute the EM potentials \textbf{A} and \phi in the Lorenz gauge. The resulting scheme is accurate, time and memory efficient, and scalable to large problems. The dispersion relation and the stability condition of this new scheme are derived and thoroughly discussed.

Moreover, the QM part utilizes a novel leapfrog method where the real and imaginary parts of the wave function are alternately updated. The method includes the time-dependent vector and scalar potentials via minimal coupling and uses a sixth-order accurate spatial discretization. The stability of this novel scheme is rigorously proven and discussed by taking the spatial variations of the potentials into account. It is shown that the stability of the scheme does not depend on the spatial gradients of the potentials, but only on their magnitude.

The remainder of this paper is organized as follows. In Section II, we present our new ADHIE method for the elec-
where \( \alpha \) is a tunable parameter, \( \Delta t \) is the time step, \( \hat{e} \) and \( \hat{h} \) are the scaled electric and magnetic field

\[
\hat{e} = \begin{bmatrix} \delta_x \otimes I_{m_y} \otimes I_{m_z} \\ \delta_y \otimes I_{m_y} \otimes I_{m_z} \\ \delta_z \otimes I_{m_y} \otimes I_{m_z} \end{bmatrix} \begin{bmatrix} e_x \\ e_y \\ e_z \end{bmatrix},
\]

\[
\hat{h} = \begin{bmatrix} \delta_x \otimes I_{n_y} \otimes I_{n_z} \\ \delta_y \otimes I_{n_y} \otimes I_{n_z} \\ \delta_z \otimes I_{n_y} \otimes I_{n_z} \end{bmatrix} \begin{bmatrix} h_x \\ h_y \\ h_z \end{bmatrix},
\]

where \( \otimes \) indicates the Kronecker product, and with \( \delta_u \) and \( \delta^*_u \), the diagonal matrices containing the \( n_u \) primary and \( n_d \) dual spatial steps in the \( u \)-direction, \( \Delta u_i \) and \( \Delta u_i^* \), respectively. The source term \( \hat{s} \) is given by

\[
\hat{s} = -\begin{bmatrix} \delta_x \otimes \delta_y^* \otimes \delta_z^* \\ \delta_y \otimes \delta_y^* \otimes \delta_z^* \\ \delta_z \otimes \delta_y^* \otimes \delta_z^* \end{bmatrix} \begin{bmatrix} j_x \\ j_y \\ j_z \end{bmatrix}.
\]

The dimensionless curl matrix \( C \) is given by

\[
C = \begin{bmatrix} 0 & -I_{n_y} \otimes I_{n_z} \otimes D_{x} & I_{n_y} \otimes D_{y} \otimes I_{n_z} & -D_{z} \otimes I_{n_y} \otimes I_{n_z} \\ I_{n_x} \otimes I_{n_y} \otimes D_{z} & -I_{n_x} \otimes D_{y} \otimes I_{n_z} & D_{z} \otimes I_{n_y} \otimes I_{n_z} \end{bmatrix}. \tag{9}
\]

with the central-difference matrices \( D_u \)

\[
D_u = \begin{bmatrix} -1 & 1 & \cdots & -1 & 1 \\ -1 & 1 & \cdots & -1 & 1 \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ -1 & 1 & \cdots & -1 & 1 \\ 1 & -1 & \cdots & 1 & -1 \end{bmatrix}_{m_u \times n_u}, \tag{10}
\]

The curl splitting matrices \( C_1 \) and \( C_2 \) are determined based on the desired local implicitization and time step. A more detailed explanation on how the local implicitization is actualized, is presented in Appendix A or in [8]. The material matrices \( M_\epsilon \) and \( M_\mu \) are

\[
M_\mu = M \begin{bmatrix} \mu \end{bmatrix}, \quad M_\epsilon = M^* \begin{bmatrix} \epsilon \end{bmatrix}, \tag{11}
\]

where the diagonal matrices \( \begin{bmatrix} \epsilon \end{bmatrix} \) and \( \begin{bmatrix} \mu \end{bmatrix} \) contain the correctly averaged permittivity \( \epsilon \) and permeability \( \mu \). The matrices \( M \) and \( M^* \) are given by

\[
M = \begin{bmatrix} (\delta_u^*)^{-1} \otimes \delta_y \otimes \delta_z & \delta_x \otimes (\delta_u^*)^{-1} \otimes \delta_z & \delta_x \otimes \delta_y \otimes (\delta_u^*)^{-1} \\ (\delta_u^{-1}) \otimes \delta_y^* \otimes \delta_z^* & \delta_y \otimes (\delta_u^{-1}) \otimes \delta_z^* & \delta_y \otimes \delta_y^* \otimes (\delta_u^{-1}) \\ (\delta_u^{-1}) \otimes \delta_y^* \otimes \delta_z^* & \delta_y \otimes (\delta_u^{-1}) \otimes \delta_z^* & \delta_y \otimes \delta_y^* \otimes (\delta_u^{-1}) \end{bmatrix}, \tag{12a}
\]

\[
M^* = \begin{bmatrix} (\delta_u^*)^{-1} \otimes \delta_y^* \otimes \delta_z^* & \delta_x \otimes (\delta_u^*)^{-1} \otimes \delta_z^* & \delta_x \otimes \delta_y \otimes (\delta_u^*)^{-1} \\ (\delta_u^{-1}) \otimes \delta_y^* \otimes \delta_z^* & \delta_y \otimes (\delta_u^{-1}) \otimes \delta_z^* & \delta_y \otimes \delta_y^* \otimes (\delta_u^{-1}) \\ (\delta_u^{-1}) \otimes \delta_y^* \otimes \delta_z^* & \delta_y \otimes (\delta_u^{-1}) \otimes \delta_z^* & \delta_y \otimes \delta_y^* \otimes (\delta_u^{-1}) \end{bmatrix}. \tag{12b}
\]

In [8], it is proven that the ADHIE scheme (5) is stable for

\[
\Delta t < \frac{2(1 - \alpha^2)}{\left\| M_\epsilon^{-\frac{1}{2}} C_0 M_\mu^{-\frac{1}{2}} \right\|_2} \tag{13}
\]

where \( C = C_0 + C_1 + C_2 \) and \( \| \cdot \|_2 \) is the matrix 2-norm.

**B. Electromagnetic potentials**

The electromagnetic potentials \( \phi \) and \( A \) in the Lorenz gauge satisfy

\[
E = -\nabla \phi - \frac{\partial A}{\partial t} \quad \text{and} \quad \nabla \cdot A + \frac{1}{c^2} \frac{\partial \phi}{\partial t} = 0, \tag{14}
\]

where \( c \) is the speed of light in vacuum. In the advocated modeling formalism, we propose to compute the potentials starting from the knowledge of the electric field and (14). Thereto, first, the EM vector potential \( A \) is discretized on the...
same spatial positions as the electric field but on half-integer
time steps resulting in
\[
a_i^{n+\frac{1}{2}} = \begin{bmatrix} a_x^{n+\frac{1}{2}} \\ a_y^{n+\frac{1}{2}} \\ a_z^{n+\frac{1}{2}} \end{bmatrix},
\]
and the scalar potential \( \phi \) is discretized on the \( n_{\phi} = m_x m_y m_z \)
internal nodes of the grid, resulting in \( \Phi \). The resulting
discretization is again shown in Fig. 1.

Second, (14) is discretized into a novel ADHIE-like scheme as follows
\[
\begin{bmatrix} I_{n_x} & 0 \\ c \Delta t D^T \end{bmatrix} \begin{bmatrix} a_i^{n+\frac{1}{2}} \\ \phi^{n+1} \end{bmatrix} = \begin{bmatrix} I_{n_x} & c \Delta t D^T \end{bmatrix} \begin{bmatrix} a_i^{n-\frac{1}{2}} \\ \phi^n \end{bmatrix} - \begin{bmatrix} e^n \\ 0 \end{bmatrix}
\]
(16)
where
\[
G = G_x \otimes G_y \otimes G_z
\]
(17)
with
\[
G_u = \left( I_{m_u} + \frac{c^2 \Delta t^2}{4 \alpha^2} \delta_u^{-1} D_u^*(1 - P_u) \delta_u^{-1} D_u^T \right)
\]
for \( u \in \{x,y,z\} \),
\[
D = \begin{bmatrix} D_1 & D_2 & D_3 \end{bmatrix}
\]
(19a)
\[
D^* = \begin{bmatrix} D_1^* & D_2^* & D_3^* \end{bmatrix}
\]
(19b)
with
\[
D_1 = D_2 \delta_x^{-1} \otimes I_{m_y} \otimes I_{m_z},
\]
\[
D_2 = I_{m_x} \otimes D_y \delta_y^{-1} \otimes I_{m_z},
\]
\[
D_3 = I_{m_x} \otimes I_{m_y} \otimes D_z \delta_z^{-1},
\]
\[
D_1^* = D_2 \delta_x^{-1} \otimes I_{m_y} \otimes I_{m_z},
\]
\[
D_2^* = I_{m_x} \otimes D_y \delta_y^{-1} \otimes I_{m_z},
\]
\[
D_3^* = I_{m_x} \otimes I_{m_y} \otimes D_z \delta_z^{-1} D_z.
\]
(20)

The diagonal and idempotent projection matrix \( P_u \in \mathbb{R}^{n_{u} \times n_{u}} \)
contains only ones or zeros and determines the local impliciti-
tization in the \( u \)-direction as
\[
[P_u]_{i,i} = p_{u,i} = \begin{cases} 0, & \text{if } \Delta u_i \text{ should be implicit,} \\ 1, & \text{if } \Delta u_i \text{ should be explicit.} \end{cases}
\]
(21)

A fully explicit scheme, as the one presented in [22], is retrieved when \( [P_u]_{i,i} = 1 \) for all \( u \) and \( i \). In contrast, to
implicitize, e.g., the \( x \)-direction completely, \( [P_x]_{i,i} \) is set equal to
0 for all \( i = 1, \ldots, n_x \). As a result, the matrix \( G = G_x \otimes I_{m_y} \otimes I_{m_z} \)
has to be inverted, only contains spatial cells in the \( x \)-direction. The parameter \( \alpha \) in (18) is again a tunable parameter. However, it will be shown in Section II-C
that, in contrast to the ADHIE method for \( E \) and \( H \) [8], it can be chosen arbitrarily close to one, drastically reducing the
splitting error. To solve (16) efficiently, the update for \( \phi \) is split in several steps given by
\[
(G_x \otimes I_{m_y} \otimes I_{m_z}) \phi_1^{(1)} = -c^2 \Delta t D^* a_i^{n+\frac{1}{2}}
\]
(22a)
\[
(I_{m_x} \otimes G_y \otimes I_{m_z}) \phi_2^{(2)} = \phi_1^{(1)}
\]
(22b)
\[
(I_{m_x} \otimes I_{m_y} \otimes G_z) \phi_3^{(3)} = \phi_2^{(2)}
\]
(22c)
\[
\phi_i^{n+1} = \phi_i^n + \phi_3^{(3)}
\]
(22d)

Consequently, the system can be solved using the tridiagonal
matrix algorithm as explained in [17], such that the complexity
remains of linear order \( \mathcal{O}(n) \) [33]. In Appendix B, the update
scheme (16) is given in a more accessible scalar notation.

C. Stability

We now present the stability of (16) with \( e = 0 \), to then
prove the stability of the complete \( (e,h,a,\phi) \) system. First,
the system (16) is transformed using the transformation matrix
\[
Q = \begin{bmatrix} \delta_x \otimes \delta_y \otimes \delta_z \\ \delta_x \otimes \delta_y \otimes \delta_z \\ \delta_x \otimes \delta_y \otimes \delta_z \end{bmatrix},
\]
(23)
to
\[
\begin{bmatrix} I_{n_x} & 0 \\ \tilde{D} & \tilde{G} \end{bmatrix} \begin{bmatrix} \tilde{a}^{n+\frac{1}{2}} \\ \tilde{\phi}^{n+1} \end{bmatrix} = \begin{bmatrix} I_{n_x} & \tilde{D}^T \end{bmatrix} \begin{bmatrix} \tilde{a}^{n-\frac{1}{2}} \\ \tilde{\phi}^n \end{bmatrix},
\]
(24)
where
\[
\tilde{a} = Q^\frac{1}{2} \begin{bmatrix} a \\ \chi \end{bmatrix},
\]
(25)
\[
\tilde{G} = \bigotimes_{u \in \{x,y,z\}} \left( I_{m_u} + \frac{1}{4 \alpha^2} \tilde{D}_u S_u \tilde{D}_u^T \right),
\]
(26)
\[
S_u = I_{n_u} - P_u, \quad \text{for } u \in \{x,y,z\},
\]
(27)
\[
\tilde{D}_u = c \Delta t \delta_u^{-2} D_u \delta_u^{-\frac{1}{2}}, \quad \text{for } u \in \{x,y,z\},
\]
(28)
with
\[
\tilde{D} = c \Delta t \left[ \delta_x \otimes \delta_y \otimes \delta_z \right]^{-\frac{1}{2}} D \left[ \delta_x \otimes \delta_y \otimes \delta_z \right]^{-\frac{1}{2}}
\]
(29)

Second, the update equation (24) is recast as
\[
(E + F) \begin{bmatrix} \tilde{a}^{n+\frac{1}{2}} \\ \tilde{\phi}^{n+1} \end{bmatrix} = (E - F) \begin{bmatrix} \tilde{a}^{n-\frac{1}{2}} \\ \tilde{\phi}^n \end{bmatrix}
\]
(30)
with
\[
E = \begin{bmatrix} I_{n_x} & \frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & 0 \end{bmatrix}, \quad \text{and } F = \begin{bmatrix} 0 & -\frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & 0 \end{bmatrix}
\]
(31)

Next, according to [34], and since \( E \) is real and symmetric and \( F \) real, the system (30) is stable if \( F + F^T \) is positive
semidefinite and \( E \) is positive definite. It is clear that \( F + F^T \)
positive semidefinite since it is the zero matrix. Matrix \( E \)
can be split as
\[
E = E' + \begin{bmatrix} 0 & 0 \\ 0 & R \end{bmatrix}
\]
(32)
where
\[
E' = \begin{bmatrix} I_{n_x} & \frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & I_{n_x} \end{bmatrix} \begin{bmatrix} I_{n_x} & \frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & I_{n_x} \end{bmatrix} = \begin{bmatrix} I_{n_x} & \frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & I_{n_x} \end{bmatrix} \begin{bmatrix} I_{n_x} & \frac{1}{2} \tilde{D}^T \\ \frac{1}{2} \tilde{D} & I_{n_x} \end{bmatrix}
\]
(33)
with
\[
P = \begin{bmatrix} P_x \otimes I_{m_y} \otimes I_{m_z} & I_{m_x} \otimes P_y \otimes I_{m_z} & I_{m_x} \otimes I_{m_y} \otimes P_z \end{bmatrix}
\]
(34)
and
\[
R = \frac{1}{16\alpha^4} \tilde{D}_x S_x \tilde{D}_x^T \otimes \tilde{D}_y S_y \tilde{D}_y^T \otimes I_{m_x} + \frac{1}{16\alpha^4} \tilde{D}_z S_z \tilde{D}_z^T \otimes I_{m_y} \otimes \tilde{D}_z S_z^T \\
+ \frac{1}{16\alpha^4} I_{m_x} \otimes \tilde{D}_y S_y \tilde{D}_y^T \otimes \tilde{D}_z S_z \\
+ \frac{1}{64\alpha^6} \tilde{D}_x S_x \tilde{D}_x^T \otimes \tilde{D}_y S_y \tilde{D}_y^T \otimes \tilde{D}_z S_z^T.
\]
(35)

All four terms in \( R \) are positive semidefinite. E.g., for the first term,
\[
x^T \left( \tilde{D}_x S_x \tilde{D}_x^T \otimes \tilde{D}_y S_y \tilde{D}_y^T \otimes I_{m_x} \right) x \\
= x^T \left( \tilde{D}_x S_x \tilde{D}_x^T \otimes \tilde{D}_y S_y \tilde{I}_{m_y} \right) x \\
= \left\| (S_x \tilde{D}_x^T \otimes S_y \tilde{D}_y^T \otimes I_{m_x}) x \right\|^2 \geq 0, \quad \forall x \in \mathbb{R}^{n_x},
\]
(36)

where we have used that \( S_n \) is idempotent. Similarly, all other terms in \( R \) can be proven to be positive semidefinite, and thus, we only need to determine when \( E' \) is positive definite.

Matrix \( E' \) is recast as
\[
E' = \begin{bmatrix}
\alpha^2 (I_{n_x} - P) & \frac{1}{2} (I_{n_x} - P) \tilde{D}_x^T \\
\frac{1}{2} \tilde{D}_x (I_{n_x} - P) & \frac{1}{2} \tilde{D}_x (I_{n_x} - P) \tilde{D}_x^T \\
\end{bmatrix}
\]
(37)

The first part \( J J^T \) is positive semidefinite by construction, so we have to determine when \( E' + \) is positive definite. The eigenvalues \( \lambda \) of \( E' \) are the solutions to \( \det (E' - \lambda I_{n_x + n_x}) = 0 \). This is transformed, using the Schur complement, to \([35]\)

\[
\det \left( (1 - \lambda) I_{n_x} - \frac{1}{4} \tilde{D}_x (1 - \lambda) I_{n_x} - \alpha^2 (I_{n_x} - P) \right)^{-1} \tilde{P} \tilde{D}_x^T = 0
\]
(40)

and

\[
\det \left( (1 - \lambda) I_{n_x} - \alpha^2 (I_{n_x} - P) \right) = 0.
\]
(41)

The latter immediately yields eigenvalues
\[
\lambda = 1 \quad \text{and} \quad \lambda = 1 - \alpha^2.
\]
(42)

Eq. (40) is simplified to
\[
\det \left( (1 - \lambda) I_{n_x} - \frac{1}{4} (1 - \lambda) \tilde{D}_x \tilde{P} \tilde{D}_x^T \right) = 0
\]
(43)

As such, the eigenvalues \( \lambda \) are related to the eigenvalues \( \sigma^2 \) of \( \tilde{D}_x \tilde{P} \tilde{D}_x^T \) by
\[
(1 - \lambda) - \frac{1}{4} \sigma^2 = 0.
\]
(44)

Note that \( \sigma \) are also the singular values of \( \tilde{D}_x \tilde{P} \), since \( P \) is idempotent. For \( E' + \) to be positive definite, we require
\[
\lambda = 1 - \frac{\sigma^2}{2} > 0.
\]
(45)

Since the matrix 2-norm is equal to the maximum singular value, we finally obtain the following stability criterion
\[
2 > \| \tilde{D}_x \tilde{P} \|_2.
\]
(46)

Eq. (46) is further simplified to
\[
\Delta t < \frac{2}{c \sqrt{\| \tilde{D}_x \tilde{P} \|_2}}.
\]
(47)

Condition (42) states that \( \alpha \) can be chosen arbitrarily close to 1 while still leaving \( E' + \) positive definite. Note that the stability criterion (47) does not depend on a splitting factor \( \alpha \) in contrast to the ADHIE method for the EM fields (13). Moreover, because \( \alpha \) can be set arbitrarily close to 1 for the potentials, the splitting error is reduced.

From (47), it is straightforward to derive a Courant-like stability criterion. Therefore, an upper bound for the 2-norm is derived:
\[
\sqrt{\| \tilde{D}_x \tilde{P} \|_2} \leq \sqrt{\| \tilde{D}_x \tilde{P} \|_\infty},
\]
(48a)

\[
= \max_{i,j,k} \left( \frac{2 p_{x,i}}{\Delta x_i^2} + \frac{p_{x,i+1}}{\Delta x_{i+1}} + \frac{p_{y,j}}{\Delta y_j^2} + \frac{p_{y,j+1}}{\Delta y_{j+1}} + \frac{2 p_{z,k}}{\Delta z_k^2} + \frac{p_{z,k+1}}{\Delta z_{k+1}} \right)^{\frac{1}{2}}.
\]
(48b)

Consequently, the system is stable if
\[
\Delta t < \frac{1}{c \sqrt{\frac{2}{\Delta x^2} + \frac{1}{\Delta y^2} + \frac{1}{\Delta z^2}}},
\]
(49)

where
\[
\frac{1}{\Delta x^2} = \max_i \left( \frac{1}{2 \Delta x_i^2} + \frac{p_{x,i+1}}{\Delta x_{i+1}} \right)
\]
(50)

and similar for \( \Delta y \) and \( \Delta z \). With this Courant-like formula, it is evident that the ADHIE scheme can remove spatial steps from the stability criterion. Typically, \( p_{u,i} \) is set equal to zero for very small steps \( \Delta t = 0 \), thus “implicitizing” them, which drastically increases the overall time step. A similar reasoning leads to a Courant-like stability criterion for the EM fields (13). Following [8] for a homogeneous background medium, we obtain:
\[
\Delta t_{ch} < \frac{(1 - \alpha^2)}{\sqrt{\frac{1}{\Delta t^2} + \frac{1}{\Delta y^2} + \frac{1}{\Delta z^2}}}.
\]
(51)
By comparing (51) with (49), we notice a reduction of the time step by a factor \((1 - \alpha^2)\).

Now, the stability of the combined system is considered. The complete \(\{e, h, a, \phi\}\) system (5)-(16) is schematically written as

\[
\begin{bmatrix}
L_{11} & 0 \\
L_{21} & L_{22}
\end{bmatrix}
\begin{bmatrix}
e^n \\
h^n planner \frac{1}{2} \\
a^n planner \frac{1}{2} \\
\phi^n + 1
\end{bmatrix} = 
\begin{bmatrix}
R_{11} & 0 \\
R_{21} & R_{22}
\end{bmatrix}
\begin{bmatrix}
e^{n-1} \\
h^{n-\frac{1}{2}} \\
a^{n-\frac{1}{2}} \\
\phi^n planner \frac{1}{2}
\end{bmatrix},
\]

(52)

where – for completeness – the occurring matrices are given in Appendix C. The stability of this system is guaranteed if the iteration matrix, which is given by

\[
\begin{bmatrix}
L_{11} \cdot R_{11} & 0 \\
- L_{22} L_{22} L_{11} - R_{22}
\end{bmatrix}
\]

has a spectral radius smaller than 1. Since this matrix is lower block triangular, the eigenvalues are given by the eigenvalues of the blocks on the diagonals. Note that these blocks \(L_{22}^{-1} R_{11}\) and \(L_{22}^{-1} R_{22}\) are the iteration matrices of the \(\{e, h\}\) system (5) and \((a, \phi)\) system (16), respectively. The stability of the complete \(\{e, h, a, \phi\}\) is thus guaranteed by guaranteeing the stability of the separate systems by imposing (13) and (47).

D. Dispersion

The accuracy of the novel ADHIE scheme for the EM potentials \(A\) and \(\phi\) is investigated by assessing the numerical dispersion. As a dispersion analysis requires constant coefficients, the grid is assumed uniform and we introduce the coefficients

\[
p_u = \begin{cases} 
0 & \text{for } u\text{-direction implicit} \\
1 & \text{for } u\text{-direction explicit} 
\end{cases}, \text{ for } u \in \{x, y, z\}.
\]

(54)

The elements of \(P_u\) are set equal to \(p_u\), i.e., \([P_u]_{i,i} = p_u\) for all \(i\). For example, \(p_x = 0, p_y = 1, \) and \(p_z = 1\) means that the \(x\)-direction is fully implicit while both the \(y\)- and \(z\)-directions are treated explicitly. We also introduce the central finite-difference notations

\[
\delta_t f^n_{l,j,k} = \frac{f_{l,j,k}^{n+\frac{1}{2}} - f_{l,j,k}^{n-\frac{1}{2}}}{\Delta t}, \\
\delta_x h^n_{l,j,k} = \frac{h_{l+\frac{1}{2},j,k}^n - h_{l-\frac{1}{2},j,k}^n}{\Delta x}, \\
\delta_x h^n_{l,j,k} = \frac{h_{l+\frac{1}{2},j,k}^n - h_{l-\frac{1}{2},j,k}^n}{\Delta y}, \\
\delta_{x,y} f^n_{l,j,k} = \frac{f_{l+1,j,k}^n - 2f_{l,j,k}^n + f_{l-1,j,k}^n}{\Delta x^2}.
\]

(55a, 55b, 55c)

As such, we can rewrite the sourceless ADI equations (16) with full implicitization in an arbitrary direction as

\[
\delta_t A_x^n_{l+\frac{1}{2},j,k} + \delta_x \phi^n_{l+\frac{1}{2},j,k} = 0,
\]

(56a)

\[
\delta_t A_y^n_{l,j+\frac{1}{2},k} + \delta_y \phi^n_{l,j+\frac{1}{2},k} = 0,
\]

(56b)

\[
\delta_t A_z^n_{l,j,k+\frac{1}{2}} + \delta_z \phi^n_{l,j,k+\frac{1}{2}} = 0,
\]

(56c)

\[
\frac{1}{c^2} \sum_u (1 + (1 - p_u)c^2 \Delta t^2 \delta_{uu}) \delta_x \phi^n_{l,j,k} + \sum_u \delta_x A_u^n_{l,j,k} = 0.
\]

(56d)

Substituting plane wave solutions of the form

\[
f^n_{l,j,k} = f_0 e^{i(\omega(n\Delta t - k_x x - k_y y - k_z z)}
\]

(57)

for \(A_x^n_{l+1/2,j,k}, A_y^n_{l,j+1/2,k}, A_z^n_{l,j,k+1/2}\), and \(\phi^n_{l,j,k+1/2}\), immediately yields

\[
\begin{bmatrix}
s_t \\
s_t \\
-s_z
\end{bmatrix}
\begin{bmatrix}
A_{0,x} \\
A_{0,y} \\
A_{0,z} \\
\phi_0
\end{bmatrix} = 
\begin{bmatrix}
0 \\
0 \\
0
\end{bmatrix}
\]

(58)

where

\[
s_t = \frac{1}{\Delta t} \sin \left(\frac{\omega \Delta t}{2}\right), \quad \sin \left(\frac{\omega \Delta t}{2}\right),
\]

(59a, 59b)

and

\[
t_u = 1 + (1 - p_u)c^2 \Delta t^2 \Delta \phi^2
\]

(59c)

By setting the determinant of the system matrix equal to zero, we find the dispersion relation

\[
\frac{1}{c^2} s_t^2 = s_x^2 + s_y^2 + s_z^2
\]

(60)

In the limit of \(\Delta t, \Delta x, \Delta y, \Delta z \to 0\), and since

\[
\lim_{\Delta u \to 0} \frac{1}{\Delta t} \sin \left(\frac{\omega \Delta t}{2}\right) = \frac{\omega}{2},
\]

(61)

and, similarly,

\[
\lim_{\Delta t \to 0} \sin \left(\frac{\omega \Delta t}{2}\right) = \omega
\]

(62)

the exact dispersion relation

\[
\omega^2 = c^2 (k_x^2 + k_y^2 + k_z^2)
\]

(63)

is retrieved.

The numerical phase error (NPE) introduced by the discretization is expressed as

\[
\text{NPE} (%) = 100 \left| \frac{\hat{k} - k_0}{k_0} \right|
\]

(64)

where \(k_0 = \omega/c\) and \(\hat{k}\) is the magnitude of the numerical wave vector calculated by numerically solving (60) after conversion to spherical coordinates. The NPE for a plane wave traveling in the first quadrant of the \(x-y\)-plane, i.e., making an angle \(\phi\) (\(0 \leq \phi \leq 90^\circ\)) with the \(x\)-axis, is shown in Fig. 2. We compare the fully explicit method to the fully implicit ADI scheme and to the hybrid implicit-explicit method with implicitization in the \(y\)-direction.
for the ADI scheme and the ADHIE-\(y\) scheme resulting in NPE = 0.110 \%.

In Fig. 2b, an unequal discretization of \(\Delta x = \Delta z = \lambda/20\), and \(\Delta y = \lambda/50\) is considered. The explicit scheme uses the Courant limit \(\Delta t_{\text{CFL}}\) for the new discretization, while the ADI and ADHIE-\(y\) schemes go beyond the Courant limit and use \(\Delta t = 1/(c\sqrt{1/\Delta x^2 + 1/\Delta z^2}) = 2.03\Delta t_{\text{CFL}}\). It is clear that the explicit scheme has again a very small error in the \(y\)-direction due to the fine sampling. Still, the error for the ADHIE-\(y\) scheme is well within acceptable limits (NPE \(\leq 0.48\%\)). Moreover, the error in the \(x\)-direction for the ADHIE-\(y\) scheme actually becomes smaller than for the explicit scheme. This is because the larger time step is closer to the 1-D magic time step \(\Delta t = \Delta x/c\) which is unstable for the fully explicit scheme. Furthermore, while the ADI scheme supports even larger time steps (beyond the magic time step), these will decrease the accuracy as there are too little temporal samples. In conclusion, the numerical dispersion relation suggests that in multiscale environments the ADHIE method is more efficient than either ADI or the explicit scheme while also having a low numerical error.

III. THE SCHRÖDINGER AND KOHN-SHAM EQUATIONS

In this section, we propose a novel method to solve the time-dependent Schrödinger or Kohn-Sham equations. Since, formally, the Kohn-Sham equation corresponds to many single-particle Schrödinger equations but with self-consistently determined potentials, the numerical time propagation of a Kohn-Sham orbital is identical to that of a single-particle wave function [36]. The differences between single-particle and many-particle systems will be addressed in Section V-B.

The minimally-coupled single-particle Schrödinger equation is

\[
\hat{H}\psi = \frac{1}{2m}(-\hbar\nabla - qA)^2 \psi + (v + q\phi)\psi,
\]  
where \(\psi\) is the wave function, \(\hat{H}\) is the Hamiltonian operator and \(v\) is a static background potential.

To construct the novel, proposed scheme for solving this equation (65), we start from the higher-order scheme [14], and extend it to include an arbitrary time-dependent vector potential \(A\) and scalar potential \(\phi\), and make it 6th-order accurate in space. Therefore, the wave function is split in its real and imaginary parts as \(\psi = r + \text{ip}\), yielding two coupled equations

\[
\begin{align*}
\hbar \frac{\partial r}{\partial t} & = -\hat{H}_0 r + \hat{H}_1 s, \\
\hbar \frac{\partial s}{\partial t} & = -\hat{H}_0 s + \hat{H}_1 r,
\end{align*}
\]

where we split the Hamiltonian \(\hat{H}\) into its real and imaginary part as \(\hat{H} = \hat{H}_0 + \text{j}\hat{H}_1\), with

\[
\begin{align*}
\hat{H}_0 & = -\frac{\hbar^2}{2m} \nabla^2 + v + \frac{q^2}{2m} A^2 + q\phi, \\
\hat{H}_1 & = \frac{q\hbar}{2m} (2A \cdot \nabla + (\nabla \cdot A)).
\end{align*}
\]

As such the Hermitian operator \(\hat{H}\) is also decomposed into a symmetric part \(\hat{H}_0\) and a skew-symmetric part \(\hat{H}_1\). The temporal discretization leveraging second-order accurate central
Around where (row-major vectorized vector $f$ where with coefficients differences and averages yields

$$\left(1 - \frac{\Delta t}{2\hbar} \hat{H}_1 \right)^{n-\frac{1}{2}} |s|^n = \left(1 + \frac{\Delta t}{2\hbar} \hat{H}_1 \right)^{n-\frac{1}{2}} |s|^{n-1}$$

(69a)

$$- \Delta t \hbar \hat{H}_0 |n-\frac{1}{2}r|^n$$

(69b)

The operator $\hat{H}_0$ is further split into $\hat{H}_0 = \hat{H}_s + \hat{H}_d$, where

$$\hat{H}_s = -\frac{\hbar^2}{2m} \nabla^2 + v$$

(70)

is static and

$$\hat{H}_d = \frac{q^2}{2m} A^2 + q\phi$$

(71)

can vary as a function of time. An alternative derivation of (69), based on the well known exponential mid-point rule [37], is given in Appendix D.

The wave function is discretized on the $n_\phi = n_\psi = m_x m_y m_z$ internal nodes of a uniform grid. The update equations can now be written concisely in matrix form as

$$\begin{bmatrix} I - \frac{\Delta t}{2\hbar} H_1 \hat{n}^{-\frac{1}{2}} & 0 \\ -\Delta t \hbar H_0 \hat{n}^{-\frac{1}{2}} & I - \frac{\Delta t}{2\hbar} H_1 \hat{n}^{-\frac{1}{2}} \end{bmatrix} \begin{bmatrix} |s|^{n} \\ |r|^{n-\frac{1}{2}} \end{bmatrix} = \begin{bmatrix} I + \frac{\Delta t}{2\hbar} H_1 \hat{n}^{-\frac{1}{2}} & -\Delta t \hbar H_0 \hat{n}^{-\frac{1}{2}} \\ 0 & I + \frac{\Delta t}{2\hbar} H_1 \hat{n}^{-\frac{1}{2}} \end{bmatrix} \begin{bmatrix} |s|^{n-1} \\ |r|^{n-\frac{1}{2}} \end{bmatrix}$$

(72)

The operators $\hat{H}_0$, $\hat{H}_1$, $\hat{H}_s$, and $\hat{H}_d$ are replaced by their discrete counterparts, i.e., the matrices $H_0, H_1, H_s, and H_d$, respectively. We introduce the notation $f \mid_{i,j,k}$ and $f \mid_{i,j,k}$, where $f \mid_{i,j,k}$ is the function evaluated at position $(x_i, y_j, z_k)$, while $f \mid_{i,j,k}$ is the $((m_y i + j)m_z + k)$-th element from the row-major vectorized vector $f$. Using this notation, the effect of $H_d$ on $f$, where $f$ can be $r$ or $s$, is given by

$$(H_d f) \mid_{i,j,k} = \frac{q^2}{2m} \left( (M_x a_x) \mid_{i,j,k} \right)^2 + \left( (M_y a_y) \mid_{i,j,k} \right)^2 + \left( M_z a_z \right) \mid_{i,j,k} f \mid_{i,j,k}$$

(73)

where $(M_x a_x) \mid_{i,j,k}$ is the sixth-order accurate average of $A_x$ around $(x_i, y_j, z_k)$, i.e.,

$$(M_x a_x) \mid_{i,j,k} = \sum_{m=-\frac{5}{2}}^{\frac{5}{2}} a^x_m A_x \mid_{i+m,j,k},$$

(74)

with coefficients

$$a^x_{-\frac{5}{2}} = a^x_{\frac{5}{2}} = \frac{3}{256}$$

(75a)

$$a^x_{-\frac{3}{2}} = a^x_{\frac{3}{2}} = \frac{25}{256}$$

(75b)

$$a^x_{-\frac{1}{2}} = a^x_{\frac{1}{2}} = \frac{75}{128}$$

(75c)

The averages in the $y$- and $z$-directions are calculated similarly.

Likewise, $H_s f$ is given by

$$(H_s f) \mid_{i,j,k} = -\frac{\hbar^2}{2m} (L f) \mid_{i,j,k} + v \mid_{i,j,k} f \mid_{i,j,k}$$

(76)

where the matrix $L$ leverages the well known sixth-order accurate discretization for the Laplacian term as used in, e.g., [38]. For the matrix $H_1$, we use

$$(H_1 f) \mid_{i,j,k} = (H_{1}^x f) \mid_{i,j,k} + (H_{1}^y f) \mid_{i,j,k} + (H_{1}^z f) \mid_{i,j,k},$$

(77)

where

$$(H_{1}^x f) \mid_{i,j,k} = \frac{9}{640\Delta x} \left( h_{i+3,j,k} f_{i+3,j,k} + h_{i+2,j,k} f_{i+2,j,k} + h_{i+1,j,k} f_{i+1,j,k} + h_{i-1,j,k} f_{i-1,j,k} + h_{i-2,j,k} f_{i-2,j,k} + h_{i-3,j,k} f_{i-3,j,k} \right),$$

(78)

with coefficients

$$h_{1,3} \mid_{i,j,k} = \frac{10}{27} A_x \mid_{i+\frac{3}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k},$$

(79a)

$$h_{1,2} \mid_{i,j,k} = -10 \left( A_x \mid_{i+\frac{3}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k} \right) - \frac{2}{3} \left( A_x \mid_{i+\frac{1}{2},j,k} + A_x \mid_{i+\frac{3}{2},j,k} \right),$$

(79b)

$$h_{1,\frac{1}{2}} \mid_{i,j,k} = \frac{290}{3} A_x \mid_{i+\frac{3}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k}$$

(79c)

$$+ 5 \left( A_x \mid_{i+\frac{3}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k} \right);$$

$$h_{1,-\frac{1}{2}} \mid_{i,j,k} = -\frac{290}{3} A_x \mid_{i-\frac{3}{2},j,k} - 5 \left( A_x \mid_{i-\frac{3}{2},j,k} + A_x \mid_{i-\frac{1}{2},j,k} \right);$$

(79d)

$$h_{1,-\frac{3}{2}} \mid_{i,j,k} = 10 \left( A_x \mid_{i-\frac{3}{2},j,k} + A_x \mid_{i-\frac{1}{2},j,k} \right) + \frac{2}{3} \left( A_x \mid_{i+\frac{3}{2},j,k} + A_x \mid_{i+\frac{1}{2},j,k} \right);$$

(79e)

$$h_{1,-\frac{5}{2}} \mid_{i,j,k} = -\frac{10}{27} A_x \mid_{i-\frac{3}{2},j,k} - A_x \mid_{i-\frac{1}{2},j,k}.$$ (79f)

The discretizations $(H_{1}^y f) \mid_{i,j,k}$ and $(H_{1}^z f) \mid_{i,j,k}$ are determined analogously. The sixth-order accurate spatial averages and differences constitute a good balance between accuracy and efficiency [39]. A Taylor expansion of $A_x$ and $f$ around $(x_i, y_j, z_k)$ shows that this discretization (78) is 6th-order accurate $O(\Delta x^6)$. Moreover, it leverages the vector potential in a symmetric way, i.e., if $A_x \mid_{i+\frac{1}{2},j,k}$ is applied to $\psi \mid_{i+\frac{1}{2},j,k}$, then $A_x \mid_{i-\frac{1}{2},j,k}$ is applied to $\psi \mid_{i-\frac{1}{2},j,k}$. Careful investigation reveals that $H_1$ is skew-symmetric for an arbitrarily varying vector potential. And since $H_0$ is symmetric, the discretized Hamiltonian $H = H_0 + jH_1$ is Hermitian, similar to its continuous counterpart $\hat{H}$.

To analyze the stability of the scheme, we assume EM fields which are constant as a function of time. Following again
the approach from [34], the update equations (72) are cast as

\[(E + F)x|^{n+1} = (E - F)x|^{n}\]

with

\[E = \begin{bmatrix} I - \frac{\Delta t}{2\hbar} H_0 \\ -\frac{\Delta t}{2\hbar} I \end{bmatrix}, \quad F = \frac{\Delta t}{2\hbar} \begin{bmatrix} -H_1 & H_0 \\ -H_0 & -H_1 \end{bmatrix}, \quad (80)\]

and

\[x|^{n} = \begin{bmatrix} s^{n-1} \\ \rho^{n-\frac{1}{2}} \end{bmatrix}. \quad (81)\]

Since \(E\) is real symmetric and \(F\) is real, the scheme is stable if \(E\) is positive definite and \(F + F^T\) is positive semidefinite. The latter is trivially satisfied because \(F + F^T\) is the zero matrix. The former is satisfied if the time step satisfies

\[\Delta t < \frac{2\hbar}{\|H_0\|_2}. \quad (82)\]

To prove this, consider that \(E\) is positive definite if

\[x^TEx > 0 \quad \forall x \in \mathbb{R}^{2n} \setminus \{0\}. \quad (83)\]

By rewriting \(E\), the positive definiteness condition (83) yields

\[\frac{\Delta t}{2\hbar} \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \otimes H_0 \right) x > \frac{\Delta t}{2\hbar} \frac{x^TAXx}{\|x\|_2^2} < 1. \quad (84)\]

We now use the fact that the maximum eigenvalue \(\lambda_{\text{max}}\) of a symmetric matrix \(A \in \mathbb{R}^{n \times n}\) is [35, eq. 8.4.3]

\[\lambda_{\text{max}}(A) = \max_{x \in \mathbb{R}^{n \times n} \setminus \{0\}} \frac{x^TAXx}{\|x\|_2^2}. \quad (85)\]

As such, (84) yields

\[\Delta t \max_{x \in \mathbb{R}^{2n} \setminus \{0\}} \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \otimes H_0 \right) x = \max_{x \in \mathbb{R}^{2n} \setminus \{0\}} \frac{x^TAXx}{\|x\|_2^2}
= \frac{\Delta t}{2\hbar} \lambda_{\text{max}} \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \otimes H_0 \right)
= \frac{\Delta t}{2\hbar} \rho(H_0)
= \frac{\Delta t}{2\hbar} \|H_0\|_2. \quad (86a)\]

Consequently, (84) yields the stability condition (82).

In going, from (86a) to (86b), it was used that the eigenvalues of a Kronecker product are the products of the eigenvalues [35] such that the spectral radius \(\rho(H_0) = \max(|\lambda_{\text{min}}(H_0)|, |\lambda_{\text{max}}(H_0)|)\), and to go from (86b) to (86c) it was used that the matrix 2-norm of a symmetric matrix is equal to its spectral radius. Note that the stability criterion does not include any gradients of potentials but only their magnitude, which are much easier to predict. As such, the obtained criterion is easily applied in simulations.

IV. COUPLING BETWEEN MAXWELL AND SCHRODINGER

The electromagnetic fields influence the behavior of the quantum system through minimal coupling (see (65)). In turn, the quantum system acts as a source for the EM fields. From the probability current density, the electric current density is obtained as

\[j = \frac{q}{2m} (-j\hbar(\psi^\ast \nabla \psi - \psi^\ast \nabla \psi) - 2qA^\ast \psi^\ast \psi) \quad (87)\]

This is rewritten in terms of the real part \(r\) and imaginary part \(s\) of the wave function

\[j = \frac{q\hbar}{m} (r \nabla s - s \nabla r - \frac{q}{\hbar}(r^2 + s^2) A). \quad (88)\]

The spatial discretization of (88) is again performed using 6th-order accurate averages and differences. The current is also temporally discretized at half integer time steps to conform with (5). The result for the \(x\)-component reads

\[j_{x|_{i+\frac{1}{2},j,k}} = \frac{q\hbar}{m} \left( (M^x r|_{(i,j,k)}(G^x s)|_{(i,j,k)})^{n+\frac{1}{2}} - (M^x s)|_{(i,j,k)}(G^x r)|_{(i,j,k)})^{n+\frac{1}{2}} \right. \]
\[\left. - \frac{q}{\hbar}(M^x_s(r \circ r + s \circ s)|_{(i,j,k)} A_{x|_{i+\frac{1}{2},j,k}}^{n+\frac{1}{2}} \right), \quad (89)\]

where \(\circ\) is the Schur product, and \((M^x r)|_{(i,j,k)}\) and \((G^x r)|_{(i,j,k)}\) are the six-point average and gradient in the \(x\)-direction of \(r\) (similar for \(s\)) around \((x, y, z)\), respectively. These are given by

\[(M^x r)|_{(i,j,k)} = \sum_{m=-\frac{1}{2}}^{\frac{1}{2}} a^x_m r|_{i+\frac{1}{2}+m}, \quad (90)\]
\[(G^x r)|_{(i,j,k)} = \sum_{m=-\frac{1}{2}}^{\frac{1}{2}} g^x_m r|_{i+\frac{1}{2}+m}, \quad (91)\]

with the \(a^x_m\) coefficients given in (75) and

\[g^x_{\frac{1}{2}} = g^x_{\frac{1}{2}} = -\frac{3}{640\Delta x}, \quad (93a)\]
\[g^x_{\frac{1}{2}} = g^x_{\frac{1}{2}} = \frac{25}{384\Delta x}, \quad (93b)\]
\[g^x_{\frac{1}{2}} = g^x_{\frac{1}{2}} = -\frac{75}{64\Delta x}. \quad (93c)\]

The other averages and gradients in (89) are calculated in an identical manner, where – however – the imaginary part of the wave function is interpolated via

\[s^{n+\frac{1}{2}}_{i,j,k} = \frac{1}{2} \left( s^{n+1}_{i,j,k} + s^n_{i,j,k} \right). \quad (94)\]

The current densities in the \(y\)- and \(z\)-directions are computed analogously. This electric current density (89) is used as a source term in (5).

V. NUMERICAL VALIDATION

The validation examples discussed in this section were all run on an Intel(R) Xeon(R) Gold 6226 CPU @ 2.70 GHz with 512 GiB of RAM memory. The simulation methods were implemented in C code because of its low-level computations and memory compactness. All examples were run on a single core and only required a small fraction of the available memory.
A. Maxwell-Schrödinger system

As a first validation example, we will replicate the results of [22] but with the much more efficient proposed scheme. A hybrid implicit-explicit discretization will be compared to a fully explicit calculation. The fully explicit calculation is nearly identical to [22]. Note that this example is also treated in [26] and [40].

A quasi 1-D nanotube [22], placed at \( x = y = 0 \text{ nm} \) and oriented along the \( z \)-direction, is illuminated by a \( z \)-polarized plane wave. The nanotube has an electrostatic confining potential \( v(z) \) defined by

\[
v(z) = v_0 \left( \frac{z}{z_{\text{max}}} \right)^4 ,
\]

where \( v_0 = 5 \times 10^3 \text{ eV} \) and \( z_{\text{max}} = 1.0 \text{ nm} \). The wave function \( \psi \) is discretized on a uniform grid with \( \Delta z = 0.01 \text{ nm} \) consisting of \( n_z = 100 \) cells. The nanotube is embedded in a uniform 3-D grid with identical discretization in the \( z \)-direction and \( n_x = n_y = 20 \) cells of \( \Delta x = \Delta y = 1 \text{ nm} \) in the \( x \)- and \( y \)-direction, respectively. An additional 10 layers of PML, described in Appendix E, are added in every direction. The wave function is normalized as

\[
\Delta x \Delta y \int |\psi|^2 \, dz = 1 .
\]

To transform a system in initial state \( \psi(t = 0) \) to a final state \( \psi_f \), the light pulse \( E_z^{\text{(in)}}(t) \) is generated “on the fly” based on the wave function \( \psi(t) \) at that time \( t \). According to [22], this is achieved with the following pulse

\[
E_z^{\text{(in)}}(t) = -2 E_0 \frac{m}{\hbar} \text{Im} \left( \langle \hat{\psi}^\prime | W q_z | \hat{\psi}^\prime \rangle \right),
\]

where

\[
W = |\psi_f \rangle \langle \psi_f | , \quad \hat{\psi}^\prime(t) = e^{-i \frac{\hbar}{m} \hat{A}_z(t) \tau} \psi(t),
\]

and

\[
E_0 = \frac{\eta}{\gamma} \exp \left( -\frac{(t - \tau) \mathbb{1}(t - \tau)}{\tau} \right).
\]

Here, \( \mathbb{1} \) is the Heaviside function, \( \gamma = 3 \text{ fs} \), \( \tau = 10 \text{ fs} \) and \( \eta = 5 \times 10^9 \text{ V/m} \). This pulse takes the EM fields generated by the moving charge inside the nanotube into account and it is generated by the following z-directed electric current sheet source which is uniform in the \( yz \)-plane and placed at \( x_s = -2 \text{ nm} \)

\[
j_z^{\text{(s)}}(x_s) = -\frac{2 E_z^{\text{(in)}}}{\Delta x z_0}.
\]

The initial state for the time-dependent simulation is \( \psi(t = 0) = \sqrt{0.9999} \psi_0 + \sqrt{0.0001} \psi_1 \), where \( \psi_0 \) is the ground state and \( \psi_1 \) is the first exited state. The ground state \( \psi_0 \) and the first excited state \( \psi_1 \) are determined by numerically calculating the eigenvectors of \( H \), where

\[
H = -\frac{\hbar^2}{2m} L_z + V.
\]

Here, \( L_z \) and \( V \) are the sixth-order accurate Laplacian matrix and discretized potential matrix as defined in Section III, but now in 1-D. The resulting wave functions are shown in Fig. 3.

Using this set-up, the goal is to design a laser pulse that transforms the initial state \( \psi(t = 0) \) to the first excited state \( \psi_1 \) and to compare the fully explicit method with the novel ADHIE method. Here, the ADHIE scheme implicizes the entire \( z \)-direction with splitting parameter \( \alpha = 0.899 \) for the \((e, \mathbf{h})\) system (5), resulting in the ADHIE-z scheme. To quantitatively compare both schemes we calculate the projections onto the ground state and first excited state as a function of time:

\[
\Omega_0(t) = |\langle \psi(t) | \psi_0 \rangle|^2, \quad \Omega_1(t) = |\langle \psi(t) | \psi_1 \rangle|^2,
\]

and compute the root-mean-square error (RMSE) for \( \Omega_1(t) \) of the ADHIE-z scheme w.r.t. the explicit scheme during the first 30 fs. The used time steps for both the explicit and ADHIE-z schemes are given in Table I. We also give the total CPU time \( T_{\text{CPU}} \), the obtained speed-up and the RMSE. Note that the time step for the ADHIE-z scheme is much closer to the maximum time step for the QM part:

\[
\Delta t_{QM} = 4.650 \times 10^{-4} \text{ fs}.
\]

In Table I, it is shown that the ADHIE-z scheme results in an 11-fold increase in efficiency for a time step that is approximately 13.5 times larger. This shows that the ADHIE method does not create much overhead. In Fig. 4, the probability density distribution \( |\psi(t)|^2 \) as a function of time is compared for the explicit scheme and the ADHIE-z scheme. It is clear that the behavior of the ADHIE-z scheme is the same despite the much larger time step. In Fig. 5, the projections \( \Omega_0(t) \) and \( \Omega_1(t) \) are shown for both schemes. It is observed that
that the explicit scheme has to use a time step \( \Delta t_{\text{exp}} = 3.335 \times 10^{-3} \text{ fs} \), while the ADHIE-z scheme can use \( \Delta t_{\text{adhie}} = 4.5 \times 10^{-4} \text{ fs} \).

\[
j \hbar \frac{\partial \psi_{l,\sigma}}{\partial t} = \frac{1}{2m} (-j\hbar \nabla - q(A_{\text{ext}} + A_{\text{ind}}[j] + A_{\text{xc},\sigma}[j_d]))^2 \psi_{l,\sigma} + (v_{\text{ext}} + v_{\text{ind}}[\rho] + v_{\text{xc},\sigma}[\rho_{\sigma}])\psi_{l,\sigma}.
\]

(104)

where \( A_{\text{ext}} \) and \( v_{\text{ext}} \) are externally applied potentials, including, e.g., background ions. The probability density \( \rho \) is given by

\[
\rho = \sum_{l,\sigma} |\psi_{l,\sigma}|^2,
\]

(105)

and \( A_{\text{xc},\sigma} \) and \( v_{\text{xc},\sigma} \) are the exchange-correlation vector and scalar potentials, respectively. The probability current density \( j \) is the sum of the probability current densities of the individual orbitals. The spin-dependent probability density \( \rho_{\sigma} \) and probability current density \( j_{\sigma} \) are defined similarly but where the sum runs only over orbitals with equal spin \( \sigma \). The induced EM potentials satisfy

\[
v_{\text{ind}} = \frac{q^2}{4\pi\epsilon} \int \frac{\rho(r',t - \frac{|r-r'|}{c})}{|r - r'|} \, dr',
\]

(106)

\[
A_{\text{ind}} = \frac{\mu_0}{4\pi} \int \frac{j(r',t - \frac{|r-r'|}{c})}{|r - r'|} \, dr'.
\]

(107)

Instead of calculating the retarded EM potentials using these integrals, as was done in [30], Maxwell’s equations are solved using the novel ADHIE method described in Section II, where \( v_{\text{ind}} = q\phi \). This circumvents saving the history dependence of the densities, thus saving memory. Moreover, the number of computations at each iteration scales linearly with the spatial domain size \( O(n_xn_yn_z) \) making it better suited for larger problems.

For the exchange and correlations potentials \( A_{\text{xc},\sigma} \) and \( v_{\text{xc},\sigma} \) we adopt the commonly used adiabatic local-density approximation (ALDA) and we also neglect the contribution of \( A_{\text{xc},\sigma} \) as was done in [30] under the assumption that its influence is inconsequential in the limit of weak magnetic fields. For simplicity, we also ignore spin such that every orbital is doubly occupied.

The addition of the exchange-correlation potential \( v_{\text{xc}} \) requires a special treatment of the density to reach a self-consistent scheme. To update \( r \) according to (69b) from \( t = (n-1/2)\Delta t \) to \( t = (n+1/2)\Delta t \), the potential \( v_{\text{xc}} \) at \( t = n\Delta t \) is required and thus also the density \( \rho_{i,j,k}^n \). The real part of the wave function \( \rho \) at time \( t = n\Delta t \) is, however, unknown and it is therefore approximated using the discretized continuity equation

\[
\rho_{i,j,k}^n = \rho_{i,j,k}^{n-1} + \frac{\Delta t}{2q} \sum_{m=-\frac{1}{2}}^{\frac{1}{2}} (g_{m,l}^x i_{|i+m,j,k}^{n-\frac{1}{2}} + g_{m,l}^y i_{j,i,j+m,k}^{n-\frac{1}{2}} + g_{m,l}^z i_{k,i,j,k+m}^{n-\frac{1}{2}})
\]

(108)

In this section, the novel ADHIE scheme for the potentials is tested for multiple electrons in a quasi 1-D nanowire while also incorporating exchange and correlation effects. This is possible within the time-dependent density-functional theory (TDDFT) framework [41], where we consider \( N \) Kohn-Sham orbitals \( \psi_{l,\sigma} \), that satisfy the single-particle Schrödinger

\[
\frac{\hbar^2}{2m} \frac{\partial^2 \psi_{l,\sigma}}{\partial x^2} + v_{\text{Kohn}}(x) \psi_{l,\sigma} = E_{l,\sigma} \psi_{l,\sigma}
\]

(30)

equation [30]

the projections are identical which is confirmed by the low RMSE\( \Omega \) in Table I. As such, we have demonstrated that the novel ADHIE scheme can be used as a much more efficient alternative to the explicit method to design a laser pulse that transforms the ground state into the first excited state.

B. Maxwell-Kohn-Sham system

In this section, the novel ADHIE scheme for the potentials is tested for multiple electrons in a quasi 1-D nanowire while also incorporating exchange and correlation effects. This is possible within the time-dependent density-functional theory (TDDFT) framework [41], where we consider \( N \) Kohn-Sham orbitals \( \psi_{l,\sigma} \), that satisfy the single-particle Schrödinger

\[
\frac{\hbar^2}{2m} \frac{\partial^2 \psi_{l,\sigma}}{\partial x^2} + v_{\text{Kohn}}(x) \psi_{l,\sigma} = E_{l,\sigma} \psi_{l,\sigma}
\]

(30)

equation [30]
where the currents are calculated with (89) and where the coefficients are given in (93). A similar approach is used half a time step earlier when updating $s$ according to (69a) from $t = (n-1)\Delta t$ to $t = n\Delta t$. As such, we now store the probability density at integer and half-integer time steps.

The model for the nanowire was developed in [42]–[44] and also recently applied in [45]. It is defined as an $N$ electron system subjected to a transverse confining harmonic potential $v_{\text{conf}}(x, y)$:

$$v_{\text{conf}}(x, y) = \frac{1}{2}m\omega_{\text{conf}}^2(x^2 + y^2).$$

(109)

Only the first transverse subband will be occupied for densities satisfying $r_s > \pi \hbar/4$ where $b^2 = \hbar/(2m\omega_{\text{conf}})$ determines the confinement and $r_s = 1/2\rho_{1d}$ is the 1-D Wigner-Seitz radius [44]. The 1-D density $\rho_{1d}$ is the 3-D density $\rho$ integrated over the transverse plane using the transverse wave function

$$\chi_{\text{tran}}(x, y) = \frac{1}{2\pi b^2}e^{-\frac{x^2 + y^2}{4b^2}}.$$ 

(110)

In [42], the resulting 1-D interelectron potential is derived as

$$v_{\text{int}}(z) = \frac{\sqrt{\pi}}{2} \frac{q^2}{4\pi e b} \exp\left(\frac{z^2}{4b^2}\right) \text{erfc}\left(\frac{z}{2b}\right),$$

(111)

where $\text{erfc}$ is the complementary error function and $\epsilon$ is the background permittivity. The Hartree potential $v_H$ – which is the static limit of $v_{\text{ind}}$ from (106) – used in density-functional theory (DFT) is then simply given by:

$$v_H[\rho_{1d}](z) = \int_{-\infty}^{\infty} v_{\text{int}}(\lvert z - z'\rvert)\rho_{1d}(z') \, dz'.$$

(112)

The corresponding ALDA parametrization for the exchange correlation potentials from [43] is used and implemented along the lines of [45].

In the longitudinal direction, the electrons are confined as in [44] by

$$v(z) = \frac{2^{\beta+1}h^2}{mL^{\beta+2}}\lvert z\rvert^\beta$$

(113)

where we choose $\beta = 6$. We also set $b = 0.1a_B$, $L = 4a_B$, $\epsilon = 11\epsilon_0$, $m = 0.063m_e$, where $m_e$ is the electron mass, $\epsilon_0$ is the permittivity of vacuum and $a_B = 4\pi \epsilon_0 h^2/mq^2$ is the effective Bohr radius. Moreover, the number of electrons $N = 6$, such that there are three doubly occupied Kohn-Sham orbitals. This set-up coincides with one of the examples in [44]. To find the ground state, the imaginary time propagation method is used [46]–[49]. For this, the wavefunctions are discretized on a uniform grid consisting of $n_z = 160$ cells with $\Delta z = 0.04a_B \approx 0.739$ nm and the sixth-order accurate finite-differences are used as described in Section III. The resulting ground state is shown in Fig. 6.

The quasi-1-D nanowire is embedded in a 3-D uniform electromagnetic grid consisting of $n_x = n_y = 20$ cells with $\Delta x = \Delta y = 6.55$ nm in the transverse directions and $n_z = 170$ cells with $\Delta z = 0.04a_B$ in the longitudinal direction. The grid is terminated by a 15 layers thick PML. The nanowire is excited with a $z$-directed uniform current sheet density in the $yz$-plane at $x_s = 32.75$ nm with temporal profile

$$j_z(x_s, t) = j_0 \sin(2\pi f_c(t - t_0)) \exp\left(-\frac{t - t_0}{2\sigma^2}\right)$$

(114)

where $j_0 = 7000\ A/m$, $f_c = 14.5 \times 10^{12}\ Hz$, $\sigma = 45.0\ fs$ and $t_0 = 200\ fs$. Since the ground state Hartree potential is merely the static contribution to the induced potential $v_{\text{ind}}$ from (106), the actual computation uses $v_{\text{int}} = q\phi + v_H[\rho_{1d}(t = 0)]$. Consequently, all fields of the $(e, h, a, \phi)$ system can be initialized to zero and updated with (5) and (16).

The $z$-directed dipole moment $p_z(t) = \int z\rho(t)\,dV$ of the nanowire and the $z$-component of the electric field at the center of the nanowire $E_{z\text{c}}$ are calculated using both the fully explicit discretization and the ADHIE method with implicitization in the $z$-direction. With these, the amplitude of the $zz$-component of the polarizability tensor $\alpha_{zz}$:

$$\alpha_{zz}(\omega) = \frac{\hat{p}_z(\omega)}{E_{z\text{c}}(\omega)},$$

(115)
and the squared amplitude of the normalized electric field at the center of the nanotube:

\[ S = \left( \frac{\hat{E}_z(\omega)}{\hat{E}_0(\omega)} \right)^2 \]  

are calculated, where \( \hat{p}_z \), \( \hat{E}_z \), and \( \hat{E}_0 \) are the Fourier transforms of the z-directed dipole moment \( p_z \), the calculated electric field, and the incident electric field, respectively. The simulation parameters and results are summarized in Table II. The ADHIE-\( z \) method supports an almost 6 times larger time step for a splitting parameter \( \alpha = 0.85 \). The achieved speed-up with the ADHIE-\( z \) method is a 4.5-fold increase. In Fig. 7, the obtained results for \( \alpha_{zz} \) and \( S \) are shown. Both for \( \alpha_{zz} \) and \( S \), the ADHIE-\( z \) method yields very similar results to the much more time-consuming fully explicit method, which is confirmed by their RMSEs given in Table II. Consequently, the novel ADHIE method is shown to be very applicable to multiscale geometries in a multiphysics TDDFT context.

VI. CONCLUSION

In this work we have presented our new framework to model combined electromagnetic (EM) and quantum mechanical (QM) problems. A novel alternating-direction implicit-explicit (ADHIE) method was developed for the EM potentials which neatly meshes with the existing ADHIE method for the EM fields. The novel method retains all the flexibility of the original method as it can apply local implicitization in preferred directions on nonuniform grids to increase the time step. Moreover, by leveraging the tridiagonal matrix algorithm, the number of calculations per time step, still scales linear, similar to the fully explicit methods. As such, the method is highly applicable for large problems in multiscale geometries. For the QM part, we substantially improved the split real and imaginary parts formulation for the time-dependent Schrödinger and Kohn-Sham equations by using sixth-order spatial differences and including arbitrary time-dependent EM potentials.

The stability of both the EM and QM parts were rigorously derived, resulting in tight upper bounds for the stability. It is noteworthy that the stability of the new ADHIE method for the potentials does not include a splitting parameter that reduces the maximum time step, as was the case for the ADHIE method for the EM fields. Also, the stability condition for the QM part does not include any gradients of the vector potential, which are hard to predict, but only its magnitude.

Numerical experiments have shown that the advocated ADHIE method increases the time step drastically both for single-particle Maxwell-Schrödinger applications and for many-particle Maxwell-Kohn-Sham systems while preserving high accuracy.

We have shown the advocated ADHIE method can be used to model hybrid EM/QM systems very efficiently. However, the ADI method upon which our hybrid method is based — while very efficient — is less accurate than the explicit method. Therefore, future work will include the investigation of alternative hybridization techniques. Additionally, the methods will be tested on more realistic devices and the electromagnetic coupling between multiple quantum systems will be investigated.

APPENDIX A

LOCAL IMPLICITIZATION

The ADHIE method generalizes the regular alternating-direction implicit (ADI) method to a hybrid implicit-explicit method. Both the regular ADI-FDTD and Yee-FDTD methods are special cases of the ADHIE-FDTD method (5). For \( C_1 = C_2 = 0 \), the conditionally stable Yee-FDTD method is...
and $\alpha = 1$, the fully implicit and unconditionally stable ADI method is found. Now, it is possible to implicitize certain directions fully or partially by modifying $C_1$ and $C_2$ to only include derivatives that have to be implicitized. For example, for partial implicitization in the $z$-direction, $C_1$ and $C_2$ are given by

$$
C_1 = \begin{bmatrix}
I_{mx} \otimes I_{ny} \otimes D_z & 0 & 0 & 0 \\
0 & D_x \otimes I_{ny} \otimes I_{nz} & 0 & 0 \\
0 & 0 & -I_{nx} \otimes I_{ny} \otimes D_z & 0 \\
-I_{mx} \otimes D_y \otimes I_{nz} & 0 & 0 & 0 \\
\end{bmatrix},
$$

$$
C_2 = \begin{bmatrix}
0 & 0 & 0 & 0 \\
-I_{nx} \otimes I_{ny} \otimes D_z(I_{nz} - P_z) & 0 & 0 & 0 \\
0 & -D_x \otimes I_{ny} \otimes I_{nz} & 0 & 0 \\
0 & 0 & -D_x \otimes I_{ny} \otimes I_{nz} & 0 \\
\end{bmatrix},
$$

where $P_z$ is defined as in (21). The stability condition (13) now depends on $C_0$, where

$$
C_0 = \begin{bmatrix}
0 & 0 & 0 & 0 \\
-I_{nx} \otimes I_{ny} \otimes D_z P_z & 0 & 0 & 0 \\
-I_{nx} \otimes D_y \otimes I_{nz} & 0 & -D_x \otimes I_{ny} \otimes I_{nz} & 0 \\
-I_{mx} \otimes I_{ny} \otimes D_z P_z & 0 & 0 & 0 \\
\end{bmatrix}.
$$

As such, it is clear that $[P_z]_{i,j,k} = 0$ removes the primary step $\Delta z_k$ from the stability criterion and that the traditional Courant limit is relaxed, without implicitizing the entire $z$-direction. For $e_x$, $e_z$, $h_x$, and $h_z$, the resulting update equations are identical to the Yee scheme – thus explicit – while for $e_y$ and $h_y$, the update equations are given by

$$
A_1 e_y^{n+\frac{1}{2}} = A_1 e_y^{n} + (I_{mx} \otimes I_{ny} \otimes (\delta^*_y)^{-1}D_z)h_x^{n-\frac{1}{2}} - ((\delta^*_y)^{-1}D_x \otimes I_{ny} \otimes I_{nz})h_x^{n-\frac{1}{2}},
$$

$$
A_2 h_y^{n+\frac{1}{2}} = A_2 h_y^{n} + (I_{mx} \otimes I_{ny} \otimes (\delta^*_z)^{-1}D^T_z) e_x^n - ((\delta^*_z)^{-1}D^T_x \otimes I_{ny} \otimes I_{nz}) e_x^n,
$$

(A.7)

with the matrices $A_1$ and $A_2$ given by (A.8)-(A.9). The diagonal material matrices $[\epsilon_x]$ and $[\mu_y]$ are the averaged permittivity and permeability for $e_x$ and $h_x$, respectively. The update equations (A.6)-(A.7) constitute tridiagonal linear systems which are smaller compared to their fully implicit counterpart. As such, the complexity is reduced w.r.t. the ADI-FDTD method while the time-step is increased w.r.t. the Yee-FDTD method, resulting in a very efficient scheme.

**APPENDIX B**

**ADHIE UPDATE EQUATIONS IN SCALAR NOTATION**

In this appendix, the novel ADHIE scheme (16) is expressed in a more straightforward scalar notation. From (16), the update equation for the $x$-component of the vector potential $A_x$ is given by:

$$
A_x^{n+\frac{1}{2},i,j,k} = A_x^{n-\frac{1}{2},i,j,k} - \frac{\Delta t}{\Delta x_i} (\phi^{n}_{i+1,j,k} - \phi^{n}_{i,j,k}) - \Delta t E_x^{n,\frac{1}{2},i,j,k}.
$$

(B.1)

Similar updates apply to $A_y$ and $A_z$. The update equations for $\phi$ are split into four steps as in (22), resulting in:

$$
\left( \zeta_x |_{i} \right) \phi^{(1)}_{i,i+1,j,k} + \left( \eta_x |_{i} \right) \phi^{(1)}_{i,j,k} + \theta_x |_{i} \phi^{(1)}_{i-1,j,k} = 0,
$$

$$
\left( \zeta_y |_{j} \right) \phi^{(2)}_{i,j+1,k} + \left( \eta_y |_{j} \right) \phi^{(2)}_{i,j,k} + \theta_y |_{j} \phi^{(2)}_{i,j-1,k} = 0,
$$

$$
\left( \zeta_z |_{k} \right) \phi^{(3)}_{i,j,k+1} + \left( \eta_z |_{k} \right) \phi^{(3)}_{i,j,k} + \theta_z |_{k} \phi^{(3)}_{i,j,k-1} = 0,
$$

$$
\phi^{n+1}_{i,j,k} = \phi^{n}_{i,j,k} + \phi^{(3)}_{i,j,k},
$$

(B.2)

where

$$
\zeta_x |_{i} = (p_x |_{i+1} - 1)\frac{c^2\Delta t^2}{4\Delta x_i \Delta x_{i+1}},
$$

$$
\theta_x |_{i} = (p_x |_{i} - 1)\frac{c^2\Delta t^2}{4\Delta x_i \Delta x_{i}},
$$

$$
\eta_x |_{i} = 1 - (\zeta_x |_{i} + \theta_x |_{i}).
$$

The update scheme (B.1)-(B.2) written in this scalar notation, makes it very evident that the update for the vector potential (B.1) is explicit and that the update for the scalar potential (B.2) constitutes at most three tridiagonal implicit systems, which can be solved very efficiently.

**APPENDIX C**

**UPDATE SCHEME FOR THE COMBINED SYSTEM**

The matrices that occur in the combined $(e, h, \alpha, \phi)$ update scheme (52) are given by:

$$
L_{11} = \begin{bmatrix}
\frac{M_h}{\Delta t} + \frac{\Delta t \epsilon^*}{\alpha} C_1 M_x^{-1} C_1^T & 0 \\
0 & \frac{M_h}{\Delta t} + \frac{\Delta t \mu^*}{\alpha} C_2^T M_x^{-1} C_2 \\
\end{bmatrix},
$$

(C.1)
$L_{21} = \begin{bmatrix} I_{n_x} & 0 \\ 0 & 0 \end{bmatrix}$,

$L_{22} = \begin{bmatrix} I_{n_x} & 0 \\ c\Delta t D^* & G \end{bmatrix}$,

$R_{11} = \begin{bmatrix} \frac{M_1}{\Delta t} + \frac{\Delta t}{2} C_1 M_{i-1} C_1^T & C \\ \frac{M_2}{\Delta t} + \frac{\Delta t}{2} C_2 M_{i+1} C_2 \\ 0 & 0 \end{bmatrix}$,

$R_{22} = \begin{bmatrix} I_{n_x} & c\Delta t D^T \\ 0 & G \end{bmatrix}$.

**APPENDIX D**

**COMPARISON WITH EXPOENTIAL MIDPOINT RULE**

In this appendix, an alternative derivation of the update equations for the Schrödinger equation (69) is given. As a starting point, we use the exponential midpoint rule. This propagator is second-order accurate in $\Delta t$, symplectic and it preserves time-reversal symmetry [50]. It is also well known in a physical-chemistry context. It states that

$$\psi|^n = \exp\left(-\frac{\Delta t}{\hbar} \hat{H}|^{n-\frac{1}{2}}\right)\psi|^{n-1}$$  \hspace{1cm} (D.1)

Now $\hat{H}$ is again split in $\hat{H}_0$ and $\hat{H}_1$ such that

$$\psi|^n = e^{\Delta t \frac{\hat{H}_1}{2}|^{n-\frac{1}{2}}} e^{-\Delta t \hat{H}_0 |^{n-\frac{1}{2}}} \psi|^{n-\frac{1}{2}}.$$  \hspace{1cm} (D.2)

This splitting introduces an error because $\hat{H}_0$ and $\hat{H}_1$ do not commute, but the error is of order $O(\Delta t^2)$. We rewrite (D.2) as a two-step procedure:

$$e^{-\frac{\Delta t}{2} \hat{H}_1 |^{n-\frac{1}{2}}} \psi|^{n-\frac{1}{2}} = e^{-\Delta t \hat{H}_0 |^{n-\frac{1}{2}}} e^{\Delta t \frac{\hat{H}_1}{2}|^{n-\frac{1}{2}}} \psi|^{n-\frac{1}{2}},$$  \hspace{1cm} (D.3)

$$e^{\frac{\Delta t}{2} \hat{H}_1 |^{n-\frac{1}{2}}} \psi|^{n-\frac{1}{2}} = e^{\Delta t \frac{\hat{H}_0}{2}|^{n-\frac{1}{2}}} e^{-\Delta t \hat{H}_1 |^{n-\frac{1}{2}}} \psi|^{n-\frac{1}{2}}.$$  \hspace{1cm} (D.4)

Taking the real part of both equations and subtracting yields

$$e^{-\frac{\Delta t}{2} \hat{H}_1 |^{n-\frac{1}{2}}} r|^n = e^{\Delta t \frac{\hat{H}_0}{2}|^{n-\frac{1}{2}}} r|^{n-1}$$

$$+ 2 \sin\left(\frac{\Delta t}{2} \hat{H}_0 |^{n-\frac{1}{2}}\right) s|^{n-\frac{1}{2}}.$$  \hspace{1cm} (D.5)

Taking the imaginary part of both equations and subtracting yields

$$e^{-\frac{\Delta t}{2} \hat{H}_1 |^{n-\frac{1}{2}}} s|^n = e^{\Delta t \frac{\hat{H}_0}{2}|^{n-\frac{1}{2}}} s|^{n-1}$$

$$- 2 \sin\left(\frac{\Delta t}{2} \hat{H}_0 |^{n-\frac{1}{2}}\right) r|^{n-\frac{1}{2}}.$$  \hspace{1cm} (D.6)

If we now make a Taylor expansion of (D.5) and (D.6), neglect terms of second order or higher in $\Delta t$, and shift (D.5) half a time step, we obtain again the proposed update scheme (69). As such, we have shown that this new scheme can be interpreted as a modification of the well-known exponential-midpoint rule and that it is second-order accurate in time.

**APPENDIX E**

**PERFECTLY MATCHED LAYER**

In this appendix, the complex frequency shifted (CFS) PML from, e.g., [51] is straightforwardly extended to the EM potentials. Therefore, a complex coordinate stretching is performed:

$$\frac{\partial}{\partial t} \rightarrow \frac{1}{s_u} \frac{\partial}{\partial t}, \quad \text{for} \quad u \in \{x, y, z\},$$  \hspace{1cm} (E.1)

where the coordinate stretching factor is defined as:

$$s_u(\omega) = \gamma_u + \frac{\sigma_u}{\gamma_u + \omega \epsilon_0}.$$  \hspace{1cm} (E.2)

By following [51] and transforming to the time domain, the following equations are obtained for the EM potentials:

$$\frac{\partial A_u}{\partial t} = -E_u - \frac{1}{\kappa_u} \left( \frac{\partial \phi}{\partial u} + \phi_u \right),$$  \hspace{1cm} (E.3)

and

$$\frac{\partial \phi}{\partial t} = -c^2 \left( \frac{1}{\kappa_x} \frac{\partial A_x}{\partial x} + \frac{1}{\kappa_y} \frac{\partial A_y}{\partial y} + \frac{1}{\kappa_z} \frac{\partial A_z}{\partial z} \right)$$

$$- c^2 \left( \frac{1}{\kappa_x} A_{xx} + \frac{1}{\kappa_y} A_{yy} + \frac{1}{\kappa_z} A_{zz} \right),$$  \hspace{1cm} (E.4)

with additional auxiliary differential equations:

$$\left( \alpha_u + \epsilon_0 \frac{\partial}{\partial t} \right) A_{uu} = -\frac{\sigma_u}{\kappa_u} \frac{\partial A_u}{\partial t},$$  \hspace{1cm} (E.5)

$$\left( \alpha_u + \epsilon_0 \frac{\partial}{\partial t} \right) \phi_u = \sigma_u \frac{\partial \phi}{\partial t},$$  \hspace{1cm} (E.6)

where

$$\alpha_u = \gamma_u + \frac{\sigma_u}{\kappa_u}.$$  \hspace{1cm} (E.7)

These are readily combined into the ADHIE update scheme resulting in the following update equations for $\phi_x$ and $A_x$:

$$\phi_{x|i + \frac{1}{2}, j, k} = a_x|i + \frac{1}{2}, j, k \phi_{x|i, j, k}$$

$$+ b_x|i + \frac{1}{2}, j, k \phi_{x|i + 1, j, k} - \phi_{x|i, j, k}$$

$$+ \Delta t E_{x|i + \frac{1}{2}, j, k} - \frac{\Delta t}{\kappa_x|i + \frac{1}{2}, j, k} \left( \phi_{x|i + 1, j, k} - \phi_{x|i, j, k} + \phi_{x|i + \frac{1}{2}, j, k} \right),$$  \hspace{1cm} (E.8)

If we now make a Taylor expansion of (D.5) and (D.6), neglect terms of second order or higher in $\Delta t$, and shift (D.5) half a time step, we obtain again the proposed update scheme (69). As such, we have shown that this new scheme can be interpreted as a modification of the well-known exponential-midpoint rule and that it is second-order accurate in time.
\[ \phi^{(0)}_{i,j,k} = -2 \Delta t \frac{\Delta x_i}{\Delta y_j} \left( A_{x}^{i+\frac{1}{2},j,k} - A_{x}^{i-\frac{1}{2},j,k} \right) + \Delta t \frac{\Delta x_i}{\Delta z_k} \left( A_{z}^{i,j+\frac{1}{2},k} - A_{z}^{i,j-\frac{1}{2},k} \right) + \Delta t \frac{\Delta y_j}{\Delta z_k} \left( A_{z}^{i,j,k+\frac{1}{2}} - A_{z}^{i,j,k-\frac{1}{2}} \right) + \Delta t \frac{\Delta z_k}{\Delta y_j} \left( A_{y}^{i,j,k+\frac{1}{2}} - A_{y}^{i,j,k-\frac{1}{2}} \right). \]

\[
\begin{align*}
\zeta_{x} & = \frac{x}{x+1} - 1, \\
\theta_{x} & = \frac{x}{x-1} - 1, \\
\eta_{x} & = 1 - (\zeta_{x} + \theta_{x}),
\end{align*}
\]

where

\[
\begin{align*}
\zeta_{x} & = (p_{x} + 1 \cdot t_{x} - 1), \\
\zeta_{y} & = (p_{y} + 1 \cdot t_{y} - 1), \\
\zeta_{z} & = (p_{z} + 1 \cdot t_{z} - 1),
\end{align*}
\]

with analogous expressions for \( A_{xy} \) and \( A_{zz} \). The coefficients \( a_{x} \) and \( b_{x} \) are given by:

\[
\begin{align*}
a_{x} & = e^{-\frac{2 \Delta t}{\alpha_{x}}} \\
b_{x} & = -\frac{\sigma_{x}}{\alpha_{x}} (1 - a_{x}).
\end{align*}
\]

In fact, these update coefficients are identical to the ones used for the EM fields [51] and are thus implemented using the same polynomial scaling for the coefficients.

REFERENCES


