Qprop Manual I

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Contents

1 Introduction

This document contains the details which were omitted in the CPC-manuscript for the sake of clarity. The short-time propagator for the case of linear polarization (propagation mode `34`) and for the more demanding case of elliptic polarization (propagation mode `44`) are discussed in great detail. Their actual implementation[1] follows exactly the lines developed in this manual. We suggest that the user reads the CPC-manuscript first.

[1]See member functions called by `propagate()` in `wavefunction.cc`.

2 Time-dependent Schrödinger and Kohn-Sham equation

The goal is to solve the time-dependent Kohn-Sham (TDKS) equation (atomic units $\hbar = |e| = m_e = 4\pi\epsilon_0 = 1$ are used, unless noted otherwise)

$$\begin{aligned} &\mathrm{i}\frac{\partial}{\partial t}\Psi_{i\sigma}(\mathbf{r},t) = \\ &\qquad \left(\frac{1}{2}[-\mathrm{i}\nabla + \mathbf{A}(t)]^2 + V(\mathbf{r}) + \mathbf{r}\cdot\mathbf{E}(t) + V_{\mathrm{ee}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]\right)\Psi_{i\sigma}(\mathbf{r},t) \end{aligned} \tag{1}$$

for the $i = 1,\ldots,N_\sigma$ Kohn-Sham (KS) orbitals $\Psi_i(\mathbf{r},t)$ of spin $\sigma = \uparrow$ and $\downarrow$, respectively. The time-dependent Schrödinger equation for a single-electron system in a laser field results from (1) by the simplification $V_{\mathrm{ee}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)] \equiv 0$ and $N_\sigma = 1$.

The two spin densities $n_\sigma(\mathbf{r},t) = n_\uparrow(\mathbf{r},t), n_\downarrow(\mathbf{r},t)$ and the total density $n(\mathbf{r},t)$ are given by

$$n_\sigma(\mathbf{r},t) = \sum_{i=1}^{N_\sigma} |\Psi_{i\sigma}(\mathbf{r},t)|^2, \qquad n(\mathbf{r},t) = n_\uparrow(\mathbf{r},t) + n_\downarrow(\mathbf{r},t). \tag{2}$$

$V(\mathbf{r})$ is the ionic background, e.g., $-Z/r$ in the case of atoms or ions of charge Z. The linearly polarized laser field is described in dipole approximation by the vector potential $\mathbf{A}(t) = A(t)\mathbf{e}_z$ or by the electric field $\mathbf{E}(t) = E(t)\mathbf{e}_z$, depending on whether the velocity gauge or the length gauge is chosen. The electron-electron interaction potential $V_{\mathrm{ee}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]$ comprises the Hartree-part $U[n(\mathbf{r},t)]$ and the exchange-correlation potential $V_{\mathrm{xc}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]$,

$$V_{\mathrm{ee}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)] = U[n(\mathbf{r},t)] + V_{\mathrm{xc}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]. \tag{3}$$

The Hartree potential

$$U[n(\mathbf{r},t)] = \int \mathrm{d}^3r'\,\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} \tag{4}$$

is due to the mutual repulsion of all the electrons. In practice, the exchange-correlation potential $V_{\mathrm{xc}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]$ has to be approximated. In time-dependent density functional studies one commonly relies on expressions for $V_{\mathrm{xc}\sigma}$ that have been successfully employed in static density functional calculations, i.e., $V_{\mathrm{xc}\sigma}$ is simply calculated with the spin densities which are present at the certain time instant.

Evaluating the square bracket in (1) a purely time-dependent term $\sim A^2(t)/2$ arises which can be transformed away by the substitution

$$\Psi_{i\sigma}(\mathbf{r},t) = \exp\left(\mathrm{i}\int\frac{A^2(t)}{2}\,\mathrm{d}t\right)\Psi'_{i\sigma}(\mathbf{r},t). \tag{5}$$

Introducing

$$\mathsf{V}_I(t) = -\mathrm{i}A(t)\frac{\partial}{\partial z} + zE(t) \tag{6}$$

one has

$$\mathrm{i}\frac{\partial}{\partial t}\Psi'_{i\sigma}(\mathbf{r},t) = \left(-\frac{1}{2}\nabla^2 + \mathsf{V}_I(t) + V(\mathbf{r}) + V_{\mathrm{ee}\sigma}[n_{\uparrow,\downarrow}(\mathbf{r},t)]\right)\Psi'_{i\sigma}(\mathbf{r},t). \tag{7}$$

We shall drop the $'$ at the KS orbitals in what follows since the phase transformation (5) has no effect on any of the potentials and observables.

For what follows we make the following assumptions and approximations:

(A) The ionic potential is assumed spherical, $V(\mathbf{r}) = V(r)$,

(B) The total electron density is assumed spin-unpolarized, i.e.,

$$n_\uparrow(\mathbf{r},t) = n_\downarrow(\mathbf{r},t), \qquad n(\mathbf{r},t) = 2n_\sigma(\mathbf{r},t), \quad \sigma = \uparrow,\downarrow, \tag{8}$$

(C) For obtaining the ground state configuration the *central field approximation* is applied, i.e., $V_{\mathrm{ee}\sigma}[n](\mathbf{r}) \to V_{\mathrm{ee}\sigma}[n](r)$,

(D) With the linearly polarized laser on, $V_{\mathrm{ee}\sigma}(\mathbf{r},t)$ is calculated up to the quadrupole term, i.e.,

$$V_{\mathrm{ee}\sigma}[n] = V^{\,0}_{\mathrm{ee}\sigma}(r,t) + V^{\,1}_{\mathrm{ee}\sigma}(r,t)\cos\vartheta + V^{\,2}_{\mathrm{ee}\sigma}(r,t)\frac{1}{2}(3\cos^2\vartheta - 1). \tag{9}$$

Note that assumption (B) is imposed only for simplicity. A generalization to spin-polarized systems would be straight forward.

2.1 Expansion in spherical harmonics

The KS orbitals $\Psi_{i\sigma}(\mathbf{r},t)$ are expanded in spherical harmonics $Y_\ell^m(\Omega)$,

$$\Psi_{i\sigma}(r,\vartheta,\varphi,t) = \frac{1}{r}\sum_{\ell=0}^{\infty}\sum_{m=-\ell}^{\ell}\Phi_{i\sigma\ell m}(r,t)Y_\ell^m(\Omega). \tag{10}$$

where Ω is the solid angle, defined by $\mathrm{d}\Omega = \sin\vartheta\,\mathrm{d}\vartheta\,\mathrm{d}\varphi$. If the initial state has a well-defined m-quantum number $m = m_i$ and the laser field is linearly polarized along the quantization axis $\mathbf{e}_z$, m_i remains a "good" quantum number and the expansion (10) simplifies to

$$\Psi_{i\sigma}(r,\vartheta,\varphi,t) = \frac{1}{r}\sum_{\ell=0}^{\infty}\Phi_{i\sigma\ell m_i}(r,t)Y_\ell^{m_i}(\Omega). \tag{11}$$

For the unperturbed atom the so-called central field approximation (CFA) is usually adopted. This leads to ground state KS orbitals of the form

$$\Psi_{i\sigma}(r,\vartheta,\varphi,0)=\frac{\Phi_{i\sigma\ell_i}(r,t)}{r}Y_{\ell_i}^{m_i}(\Omega), \tag{12}$$

having well-defined quantum numbers ℓ_i and m_i with the radial function independent of m_i. As mentioned above, during the interaction with the linearly polarized laser field in dipole approximation m_i remains a "good" quantum number, i.e., no other $m \neq m_i$ will be populated thanks to the azimuthal symmetry that is retained throughout the interaction with the laser field. However, the broken spherical symmetry introduces a mixing of angular momentum and renders the radial wavefunction dependent on m. The KS equations become

$$\begin{aligned}
\mathrm{i}\frac{\partial}{\partial t}\Phi_{i\sigma\ell m_i}(r,t) \quad = \quad & \left(-\frac{1}{2}\frac{\partial^2}{\partial r^2}+V_\ell^{\mathrm{eff}}(r)\right)\Phi_{i\ell m_i}(r,t) \\
& -\mathrm{i}A(t)r\sum_{\ell'}\langle Y_\ell^{m_i}|\cos\vartheta|Y_{\ell'}^{m_i}\rangle\frac{\partial}{\partial r}\frac{1}{r}\Phi_{i\ell' m_i}(r,t) \\
& +\mathrm{i}A(t)\sum_{\ell'}\langle Y_\ell^{m_i}|\sin\vartheta\frac{\partial}{\partial\vartheta}|Y_{\ell'}^{m_i}\rangle\frac{1}{r}\Phi_{i\ell' m_i}(r,t) \\
& +rE(t)\sum_{\ell'}\langle Y_\ell^{m_i}|\cos\vartheta|Y_{\ell'}^{m_i}\rangle\Phi_{i\ell' m_i}(r,t) \\
& +\sum_{\ell'}\langle Y_\ell^{m_i}|V_{\mathrm{ee}\sigma}[n_\sigma]+V(\mathbf{r})-V^0(r)|Y_{\ell'}^{m_i}\rangle\Phi_{i\ell' m_i}(r,t)
\end{aligned} \tag{13}$$

where we suppressed the spin index σ. In the last four lines of (13) we made use of the fact that the corresponding matrix elements contribute for $m'=m$ only. $V_\ell^{\mathrm{eff}}(r)$ is the effective ionic potential including the centrifugal barrier and the spherical part $V^0(r)$ of the ionic potential, $V_\ell^{\mathrm{eff}}(r)=V^0(r)+\frac{\ell(\ell+1)}{2r^2}$. In the forthcoming section we specialize on spherical ionic potentials, i.e., atoms, ions, or jellium clusters so that $V(\mathbf{r})-V^0(r)=0$. Moreover, in CFA $V_{\mathrm{ee}\sigma}[n_\sigma]$ is diagonal in ℓ and m. However, as the laser is switched on, the density n is not spherical any longer, as is $V_{\mathrm{ee}\sigma}[n_\sigma]$. A full multipole expansion of $V_{\mathrm{ee}\sigma}[n_\sigma]$ would lead to a densely populated matrix $\langle Y_\ell^m|V_{\mathrm{ee}\sigma}[n_\sigma]|Y_{\ell'}^m\rangle$. Hence, the efficiency of the numerical scheme rises and falls with the possibility to terminate the multipole expansion after a few terms.

Defining

$$c_{\ell m}=\sqrt{\frac{(\ell+1)^2-m^2}{(2\ell+1)(2\ell+3)}} \tag{14}$$

one can write the matrix elements

$$r\sum_{\ell'}\langle Y_\ell^{m_i}|\cos\vartheta|Y_{\ell'}^{m_i}\rangle\Phi_{i\ell' m_i} \quad = \quad r\Big(c_{\ell-1,m_i}\Phi_{i,\ell-1,m_i}+c_{\ell m_i}\Phi_{i,\ell+1,m_i}\Big), \tag{15}$$

$$r\sum_{\ell'}\langle Y_\ell^{m_i}|\cos\vartheta|Y_{\ell'}^{m_i}\rangle\frac{\partial}{\partial r}\frac{1}{r}\Phi_{i\ell' m_i} = \Big(c_{\ell-1,m_i}\frac{\partial}{\partial r}\Phi_{i,\ell-1,m_i}+c_{\ell m_i}\frac{\partial}{\partial r}\Phi_{i,\ell+1,m_i}\Big) -\Big(\frac{c_{\ell-1,m_i}}{r}\Phi_{i,\ell-1,m_i}+\frac{c_{\ell m_i}}{r}\Phi_{i,\ell+1,m_i}\Big), \quad (16)$$

and

$$\sum_{\ell'}\langle Y_\ell^{m_i}|\sin\vartheta\frac{\partial}{\partial\vartheta}|Y_{\ell'}^{m_i}\rangle\frac{1}{r}\Phi_{i\ell' m_i} \quad (17)$$
$$=\frac{1}{r}\Big((\ell-1)c_{\ell-1,m_i}\Phi_{i,\ell-1,m_i}-(\ell+2)c_{\ell m_i}\Phi_{i,\ell+1,m_i}\Big).$$

The matrix element $\langle Y_\ell^{m_i}|\cos\vartheta|Y_{\ell'}^{m_i}\rangle$ also appears when the dipole term ${V_{\mathrm{ee}}}_\sigma^1(r,t)\cos\vartheta$ is introduced into (13). The quadrupole term instead leads to

$$\frac{1}{2}\sum_{\ell'}\langle Y_\ell^{m_i}|3\cos^2\vartheta-1|Y_{\ell'}^{m_i}\rangle\Phi_{i\ell' m_i} \quad (18)$$
$$=p_{\ell m_i}\Phi_{i\ell m_i}+q_{\ell-2,m_i}\Phi_{i,\ell-2,m_i}+q_{\ell m_i}\Phi_{i,\ell+2,m_i}$$

where

$$p_{\ell m}=\frac{\ell(\ell+1)-3m^2}{(2\ell-1)(2\ell+3)}, \quad (19)$$

$$q_{\ell m}=\frac{3}{2(2\ell+3)}\sqrt{\frac{[(\ell+1)^2-m^2][(\ell+2)^2-m^2]}{(2\ell+1)(2\ell+5)}}. \quad (20)$$

Inserting (9) into the TDKS equation (13) and using (15)–(18) one obtains

$$\begin{aligned}\mathrm{i}\frac{\partial}{\partial t}\Phi_{i\ell m_i} &= \Big(-\frac{1}{2}\frac{\partial^2}{\partial r^2}+V_\ell^{\mathrm{eff}}(r)+V_{\mathrm{ee}}^0(r,t)+p_{\ell m_i}V_{\mathrm{ee}}^2(r,t)\Big)\Phi_{i\ell m_i} \quad (21)\\ &\quad -\mathrm{i}A(t)\Big(c_{\ell-1,m_i}\frac{\partial}{\partial r}\Phi_{i,\ell-1,m_i}+c_{\ell m_i}\frac{\partial}{\partial r}\Phi_{i,\ell+1,m_i}\\ &\qquad -\frac{1}{r}\ell c_{\ell-1,m_i}\Phi_{i,\ell-1,m_i}+\frac{1}{r}(\ell+1)c_{\ell m_i}\Phi_{i,\ell+1,m_i}\Big)\\ &\quad +\Big(rE(t)+V_{\mathrm{ee}}^1(r,t)\Big)\Big(c_{\ell-1,m_i}\Phi_{i,\ell-1,m_i}+c_{\ell m_i}\Phi_{i,\ell+1,m_i}\Big)\\ &\quad +V_{\mathrm{ee}}^2(r,t)\Big(q_{\ell-2,m_i}\Phi_{i,\ell-2,m_i}+q_{\ell m_i}\Phi_{i,\ell+2,m_i}\Big).\end{aligned}$$

2.2 Expansion of the Hartree potential up to the quadrupole

In the expansion (9) each term ${V_{\mathrm{ee}}}_\sigma^j$, $j=0,1,2$, consists of the Hartree part U^j and the exchange-correlation part ${V_{\mathrm{xc}}}_\sigma^j$.

With the spin density (2) written as

$$n_\sigma(\mathbf{r},t) = \frac{1}{r^2}\sum_{i=1}^{N_\sigma}\sum_{\ell\ell'}\Phi^*_{i\ell' m_i}(r,t)\Phi_{i\ell m_i}(r,t)Y_{\ell'}^{m_i *}(\Omega)Y_\ell^{m_i}(\Omega) \tag{22}$$

and the well-known identity

$$\frac{1}{|\mathbf{r}-\mathbf{r}'|} = \sum_{\ell m}\frac{4\pi}{2\ell+1}\,\frac{r_<^\ell}{r_>^{\ell+1}}\;Y_\ell^{m*}(\Omega)Y_\ell^m(\Omega'), \tag{23}$$

where $r_< = \min(r,r')$, $r_> = \max(r,r')$, it can be shown that the Hartree potential (4) is given by

$$\begin{aligned} U[n] &= 2\sum_L Y_L^0(\Omega)\sqrt{\frac{4\pi}{2L+1}}\int\!\mathrm{d}r'\,\frac{r_<^L}{r_>^{L+1}} \\ &\times\sum_{i=1}^{N_\sigma}\sum_{\ell\ell'}\Phi^*_{i\ell' m_i}(r',t)\Phi_{i\ell m_i}(r',t)\sqrt{\frac{2\ell+1}{2\ell'+1}}C^{\ell'0}_{\ell 0L0}C^{\ell' m_i}_{\ell m_i L0}. \end{aligned} \tag{24}$$

$C^{c\gamma}_{a\alpha b\beta}$ are the Clebsch-Gordan coefficients. Using the property $C^{\ell'0}_{\ell 0L0}C^{\ell' m}_{\ell mL0} = \frac{2\ell'+1}{2\ell+1}C^{\ell 0}_{\ell' 0L0}C^{\ell m}_{\ell' mL0}$ Eq. (24) can be written in the form

$$U[n] = 2\sum_L Y_L^0(\Omega)\sqrt{\frac{4\pi}{2L+1}}\int\!\mathrm{d}r'\,\frac{r_<^L}{r_>^{L+1}}\sum_{i=1}^{N_\sigma}\tilde{\Lambda}^L_{ii}(r',t) \tag{25}$$

where we introduced the entities

$$\tilde{\Lambda}^L_{ji}(r,t) = \tilde{\Lambda}^{Lm_j-m_i}_{ji}(r,t), \tag{26}$$

$$\tilde{\Lambda}^{LM}_{ji}(r,t) = \sum_{\ell\ell'}\sqrt{\frac{2\ell+1}{2\ell'+1}}C^{\ell'0}_{\ell 0L0}C^{\ell' m_j}_{\ell m_i LM}\Phi^*_{i\ell m_i}(r,t)\Phi_{j\ell' m_j}(r,t). \tag{27}$$

$\tilde{\Lambda}^{LM}_{ji}(r,t)$ is a key expression and will appear frequently in our forthcoming discussion of the KLI exchange potential.

In terms of the auxiliary entities

$$\Lambda(r,t) = 2\sum_{i=1}^{N_\sigma}\sum_\ell|\Phi_{i\ell m_i}(r,t)|^2 = 2\sum_{i=1}^{N_\sigma}\tilde{\Lambda}^0_{ii}(r,t), \tag{28}$$

$$\begin{aligned} \Theta(r,t) &= 2\sum_{i=1}^{N_\sigma}\sum_\ell\Big(c_{\ell-1,m_i}\Phi^*_{i,\ell-1,m_i} + c_{\ell m_i}\Phi^*_{i,\ell+1,m_i}\Big)\Phi_{i\ell m_i} \\ &= 2\sum_{i=1}^{N_\sigma}\tilde{\Lambda}^1_{ii}(r,t), \end{aligned} \tag{29}$$

$$
\begin{aligned}
\Xi(r,t) &= 2\sum_{i=1}^{N_\sigma}\sum_\ell \Big(p_{\ell m_i}\Phi^*_{i\ell m_i} + q_{\ell m_i}\Phi^*_{i,\ell+2,m_i} + q_{\ell-2,m_i}\Phi^*_{i,\ell-2,m_i}\Big)\Phi_{i\ell m_i} \\
&= 2\sum_{i=1}^{N_\sigma}\tilde{\Lambda}^2_{ii}(r,t)
\end{aligned} \tag{30}
$$

one obtains

$$U^0(r,t) = \int \mathrm{d}r' \, \frac{1}{r_>} \, \Lambda(r',t), \tag{31}$$

$$U^1(r,t) = \int \mathrm{d}r' \, \frac{r_<}{r_>^2} \, \Theta(r',t), \tag{32}$$

$$U^2(r,t) = \int \mathrm{d}r' \, \frac{r_<^2}{r_>^3} \, \Xi(r',t). \tag{33}$$

3 Propagation scheme for linear polarization

3.1 Breaking down the Hamiltonian

In this and the subsequent Section we essentially proceed in line with the work by H.G. Muller [Laser Physics **9**, 138 (1999)]. In matrix notation (21) may be written as

$$\mathrm{i}\partial_t\mathbf{\Phi}(r,t) = \Big(\mathsf{H}_{\mathrm{at}} + \mathsf{H}_{\mathrm{mix}} + \mathsf{H}^{(1)}_{\mathrm{ang}} + \mathsf{H}^{(2)}_{\mathrm{ang}} + \mathsf{H}^{(3)}_{\mathrm{ang}}\Big)\mathbf{\Phi}(r,t) \tag{34}$$

with

$$\mathsf{H}_{\mathrm{at}} = -\frac{1}{2}\frac{\partial^2}{\partial r^2} + \mathsf{V}^\ell_{\mathrm{eff}} + V^0_{\mathrm{ee}} + p_{\ell m}V^2_{\mathrm{ee}}. \tag{35}$$

In this Section we suppress the orbital index i of the quantum number m. It is understood that the m_i of the KS orbital on which the operator acts has to be taken. $\mathbf{\Phi}(r,t) = (\mathbf{\Phi}_{0m}(r,t), \mathbf{\Phi}_{1m}(r,t), \ldots)^\top$ is a vector in ℓ-space with the quantum number m arbitrary but fixed, and, of course, only ℓs with $\ell \geq |m|$ are allowed. The matrices $\mathsf{H}_{\mathrm{mix}}$, $\mathsf{H}^{(1)}_{\mathrm{ang}}$, $\mathsf{H}^{(2)}_{\mathrm{ang}}$, and $\mathsf{H}^{(3)}_{\mathrm{ang}}$ are

$$
\begin{aligned}
\mathsf{H}_{\mathrm{mix}} &= -\mathrm{i}A(t)\begin{pmatrix} 0 & c_{0m} & 0 & 0 & \cdots \\ c_{0m} & 0 & c_{1m} & 0 & \\ 0 & c_{1m} & 0 & c_{2m} & \\ \vdots & & c_{2m} & & \end{pmatrix}\partial_r, \\
\mathsf{H}^{(1)}_{\mathrm{ang}} &= -\mathrm{i}\frac{A(t)}{r}\begin{pmatrix} 0 & c_{0m} & 0 & 0 & \cdots \\ -c_{0m} & 0 & 2c_{1m} & 0 & \\ 0 & -2c_{1m} & 0 & 3c_{2m} & \\ \vdots & & -3c_{2m} & & \end{pmatrix},
\end{aligned}
$$

$$
\mathsf{H}^{(2)}_{\text{ang}} = \left(rE(t) + V^1_{\text{ee}}(r,t)\right) \begin{pmatrix} 0 & c_{0m} & 0 & 0 & \cdots \\ c_{0m} & 0 & c_{1m} & 0 & \\ 0 & c_{1m} & 0 & c_{2m} & \\ \vdots & & c_{2m} & & \end{pmatrix},
$$

$$
\mathsf{H}^{(3)}_{\text{ang}} = V^2_{\text{ee}}(r,t) \begin{pmatrix} 0 & 0 & q_{0m} & 0 & \cdots \\ 0 & 0 & 0 & q_{1m} & \\ q_{0m} & 0 & 0 & & \\ \vdots & q_{1m} & & & \end{pmatrix}.
$$

Each entry in the vector $\mathbf{\Phi}(r,t)$ depends on r. In the code, r-space is discretized,

$$
r_n = n\Delta r, \qquad n = 1,2,3,\ldots,N_r
$$

so that our discrete "numerical" vector is of the form

$$
\mathbf{\Psi}(t) = \left(\boxed{\Phi^1_{0m}(t),\ldots,\Phi^{N_r}_{0m}(t)}, \boxed{\Phi^1_{1m}(t),\ldots,\Phi^{N_r}_{1m}(t)}, \boxed{\ldots,\Phi^{N_r}_{L-1,m}(t)}\right)^\top,
$$

where N_r and L are the number of grid points in r- and ℓ-space, respectively. Now the different pieces of the total Hamiltonian,

$$
\mathsf{H} = \mathsf{H}_{\text{at}} + \mathsf{H}_{\text{mix}} + \mathsf{H}_{\text{ang}}, \tag{36}
$$

$$
\mathsf{H}_{\text{ang}} = \mathsf{H}^{(1,2)}_{\text{ang}} + \mathsf{H}^{(3)}_{\text{ang}}, \qquad \mathsf{H}^{(1,2)}_{\text{ang}} = \mathsf{H}^{(1)}_{\text{ang}} + \mathsf{H}^{(2)}_{\text{ang}},
$$

operating in r- and ℓ-space, will be analyzed.

H_{at} is diagonal in ℓ-space, $\mathsf{H}^{(1,2)}_{\text{ang}}$ and $\mathsf{H}^{(3)}_{\text{ang}}$ are diagonal in r-space. H_{mix} is, unfortunately, nowhere diagonal. The matrices H_{mix} and H_{ang} can be written as a sum of matrices acting in two-dimensional $\ell, \ell+1$ or $\ell, \ell+2$ subspaces only, for example,

$$
\begin{pmatrix} 0 & c_{0m} & 0 & \cdots \\ c_{0m} & 0 & c_{1m} & \\ 0 & c_{1m} & 0 & \ddots \\ \vdots & & \ddots & \end{pmatrix} = \begin{pmatrix} 0 & c_{0m} & 0 \\ c_{0m} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0\cdots \\ 0 & 0 & c_{1m} \\ 0 & c_{1m} & 0 \end{pmatrix} + \cdots
$$

One matrix in this series couples a certain angular momentum ℓ with $\ell+1$. In what follows the corresponding Hamilton operator is labeled accordingly:

$$
\mathsf{H}^{\ell m}_{\text{mix}} \begin{pmatrix} \Phi_{\ell m}(r,t) \\ \Phi_{\ell+1,m}(r,t) \end{pmatrix} = -\mathrm{i}A(t) \begin{pmatrix} 0 & c_{\ell m} \\ c_{\ell m} & 0 \end{pmatrix} \partial_r \begin{pmatrix} \Phi_{\ell m}(r,t) \\ \Phi_{\ell+1,m}(r,t) \end{pmatrix}, \tag{37}
$$

$$
\begin{aligned}
&\mathsf{H}^{(1,2)\ell m}_{\text{ang}} \begin{pmatrix} \Phi_{\ell m}(r,t) \\ \Phi_{\ell+1,m}(r,t) \end{pmatrix} \qquad\qquad (38) \\
&= \left\{ -\frac{\mathrm{i}A(t)}{r} \begin{pmatrix} 0 & t_{\ell m} \\ -t_{\ell m} & 0 \end{pmatrix} + \left(rE(t) + V^1_{\text{ee}}(r,t)\right) \begin{pmatrix} 0 & c_{\ell m} \\ c_{\ell m} & 0 \end{pmatrix} \right\} \begin{pmatrix} \Phi_{\ell m}(r,t) \\ \Phi_{\ell+1,m}(r,t) \end{pmatrix},
\end{aligned}
$$

where

$$t_{\ell m} = (\ell+1)\sqrt{\frac{(\ell+1)^2 - m^2}{(2\ell+1)(2\ell+3)}}, \tag{39}$$

and

$$\mathsf{H}_{\text{ang}}^{(3)\ \ell m}\begin{pmatrix}\Phi_{\ell m}(r,t)\\ \Phi_{\ell+2,m}(r,t)\end{pmatrix} = V_{\text{ee}}^2(r,t)\begin{pmatrix}0 & q_{\ell m}\\ q_{\ell m} & 0\end{pmatrix}\begin{pmatrix}\Phi_{\ell m}(r,t)\\ \Phi_{\ell+2,m}(r,t)\end{pmatrix}. \tag{40}$$

Spatial derivatives with respect to the radius r are present in H_{at} and H_{mix}. The Numerov and Simpson approximants to the second and first derivative of a function $f(r)$ are

$$f'' = -2\mathsf{M}_2^{-1}\Delta_2 f, \quad f' = \mathsf{M}_1^{-1}\Delta_1 f, \tag{41}$$

respectively, where $\Delta_2 f = (f_{n+1} - 2f_n + f_{n-1})/h^2$, $h = \Delta r$, $\Delta_1 f = (f_{n+1} - f_{n-1})/2h$, and

$$\mathsf{M}_2 = -\frac{1}{6}\begin{pmatrix}10 & 1 & 0 & \cdots \\ 1 & 10 & 1 & \\ 0 & 1 & 10 & 1 \\ \vdots & & & \end{pmatrix}, \quad \mathsf{M}_1 = \frac{1}{6}\begin{pmatrix}4 & 1 & 0 & \cdots \\ 1 & 4 & 1 & \\ 0 & 1 & 4 & 1 \\ \vdots & & & \ddots\end{pmatrix}. \tag{42}$$

M_2 and M_2 operate on $\mathbf{f} = (f_1, f_2, \ldots, f_{N_r})^\top$. Applied to our problem, the matrices M_1, M_2, Δ_1, Δ_2 act in r-space, i.e., on all ℓ-blocks $\boldsymbol{\Phi}_\ell(t) = (\Phi_\ell^1(t), \Phi_\ell^2(t), \ldots)^\top$ independently.

The upper left matrix elements of M_2 and Δ_2 have to be modified because (42) would imply $f''(r_0) = 0$ (note that r_0 is not part of the grid). However, for a Coulomb-potential $-Z/r$ and $\ell = 0$ at $r_0 = 0$

$$\Phi_0''(0,t) = -2Z\Phi_0'(0,t) \neq 0$$

holds. Moreover, one has to ensure the Hermiticity of $\tilde{\mathsf{M}}_2^{-1}\tilde{\Delta}_2$ in order not to destroy unitary propagation. Both may be achieved by modifying the upper left matrix elements,

$$\begin{aligned}(\tilde{\Delta}_2)_{1,1} &= -\frac{2}{h^2}\left(1 - \frac{Zh}{12 - 10Zh}\right), \\ (\tilde{\mathsf{M}}_2)_{1,1} &= -2\left(1 + \frac{h^2}{12}(\tilde{\Delta}_2)_{1,1}\right).\end{aligned}$$

The corner elements of Δ_1 and M_1 have to be modified as well because the operator $\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1$ must be anti-Hermitian for a unitary time propagation. Writing

$$\tilde{\mathsf{M}}_1 = \frac{1}{6}\begin{pmatrix}4+x & 1 & & \\ 1 & 4 & 1 & \\ & & \ddots & \\ & & 1 & 4+x'\end{pmatrix}, \quad \tilde{\Delta}_1 = \frac{1}{2h}\begin{pmatrix}y & 1 & & \\ -1 & 0 & 1 & \\ & & \ddots & \\ & & -1 & y'\end{pmatrix}$$

one finds that with $x = \sqrt{3} - 2$, $x' = y = x$ and $y' = -y$ anti-Hermiticity of $\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1$ is ensured.

To summarize, we write the different contributions to the total Hamilton as follows:

$$
\begin{aligned}
\mathsf{H} &= \mathsf{H}_{\mathrm{at}} + \sum_{\ell=0}^{L-2} \left(\mathsf{H}_{\mathrm{mix}}^{\ell m} + \mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m} + \mathsf{H}_{\mathrm{ang}}^{(3)\;\ell m} \right), && (43)\\
\mathsf{H}_{\mathrm{at}} &= \mathbf{1}_\ell \otimes \left(\tilde{\mathsf{M}}_2^{-1}\tilde{\Delta}_2 + \mathsf{V}_{\mathrm{eff}}^{\ell} + V_{\mathrm{ee}}^0 + p_{\ell m} V_{\mathrm{ee}}^2 \right), && (44)\\
\mathsf{H}_{\mathrm{mix}}^{\ell m} &= -\mathrm{i}A(t)\mathsf{L}_{\ell m} \otimes \tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1, && (45)\\
\mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m} &= -\mathrm{i}A(t)\mathsf{T}_{\ell m} \otimes \frac{1}{r_n}\mathbf{1}_r + \mathsf{L}_{\ell m} \otimes \Big(r_n E(t) + V_{\mathrm{ee}}^1(r_n, t) \Big)\mathbf{1}_r, && (46)\\
\mathsf{H}_{\mathrm{ang}}^{(3)\;\ell m} &= \mathsf{P}_{\ell m} \otimes V_{\mathrm{ee}}^2(r_n, t)\mathbf{1}_r, && (47)
\end{aligned}
$$

where $\mathbf{1}_\ell$ and $\mathbf{1}_r$ are unity matrices in ℓ- and r-space, respectively, and

$$
\mathsf{L}_{\ell m} = \begin{pmatrix} 0 & c_{\ell m} \\ c_{\ell m} & 0 \end{pmatrix}, \quad \mathsf{T}_{\ell m} = \begin{pmatrix} 0 & t_{\ell m} \\ -t_{\ell m} & 0 \end{pmatrix}, \quad \mathsf{P}_{\ell m} = \begin{pmatrix} 0 & q_{\ell m} \\ q_{\ell m} & 0 \end{pmatrix}. \qquad (48)
$$

Note that $\mathsf{L}_{\ell m}$ and $\mathsf{T}_{\ell m}$ act on $\ell, \ell+1$-sub blocks while $\mathsf{P}_{\ell m}$ acts on $\ell, \ell+2$-sub blocks.

3.2 Approximating the time evolution operator

For a sufficiently small time step

$$
\mathsf{U}(t + \Delta t, t) = \exp(-\mathrm{i}\Delta t \mathsf{H}(t + \Delta t/2))
$$

is a good approximation to the exact propagator, i.e., $\mathbf{\Psi}(t + \Delta t) = \mathsf{U}(t + \Delta t, t)\mathbf{\Psi}(t)$. With the Hamiltonian (43) an approximation, accurate up to second order in Δt, is given by

$$
\begin{aligned}
\mathsf{U}_{\mathrm{split}}(t + \Delta t, t) = & \prod_{\ell=L-3}^{0} \exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{ang}}^{(3)\;\ell m}) \\
& \times \prod_{\ell=L-2}^{0} \left(\exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m}) \exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{mix}}^{\ell m}) \right) \\
& \times \exp(-\mathrm{i}\Delta t \mathsf{H}_{\mathrm{at}}) \\
& \times \prod_{\ell=0}^{L-2} \left(\exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{mix}}^{\ell m}) \exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m}) \right) \\
& \times \prod_{\ell=0}^{L-3} \exp(-\mathrm{i}\tau \mathsf{H}_{\mathrm{ang}}^{(3)\;\ell m}) \qquad (49)
\end{aligned}
$$

with $\tau = \Delta t/2$. The unitary Crank-Nicolson (CN) approximant to $\exp(-\mathrm{i}\Delta t \mathsf{H})$ is

$$\exp(-\mathrm{i}\Delta t \mathsf{H}) = (\mathbf{1} + \mathrm{i}\Delta t \mathsf{H}/2)^{-1}(\mathbf{1} - \mathrm{i}\Delta t \mathsf{H}/2) + O(\Delta t)^3$$

so that

$$\begin{aligned} \mathsf{U}_{\mathrm{split}}(t+\Delta t, t) &\approx \mathsf{U}_{\mathrm{CN}}(t+\Delta t, t) \\ &= \prod_{\ell=L-3}^{0} \mathsf{Z}_n^{\ell m} \prod_{\ell=L-2}^{0} \left(\mathsf{R}_n^{\ell m} \left[\mathsf{X}_+^{\ell m}\right]^{-1} \mathsf{X}_-^{\ell m} \right) \mathsf{Q}_+^{-1}\, \mathsf{Q}_- \\ &\qquad \times \prod_{\ell=0}^{L-2} \left(\left[\mathsf{X}_+^{\ell m}\right]^{-1} \mathsf{X}_-^{\ell m}\, \mathsf{R}_n^{\ell m} \right) \prod_{\ell=0}^{L-3} \mathsf{Z}_n^{\ell m} \end{aligned} \tag{50}$$

where

$$\mathsf{R}_n^{\ell m} = \left(\mathbf{1} + \mathrm{i}\frac{\tau}{2} \mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m}\right)^{-1} \left(\mathbf{1} - \mathrm{i}\frac{\tau}{2} \mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m}\right), \tag{51}$$

$$\mathsf{X}_\pm^{\ell m} = \mathbf{1} \pm \mathrm{i}\frac{\tau}{2} \mathsf{H}_{\mathrm{mix}}^{\ell m}, \quad \mathsf{Q}_\pm = \mathbf{1} \pm \mathrm{i}\tau \mathsf{H}_{\mathrm{at}}, \tag{52}$$

$$\mathsf{Z}_n^{\ell m} = \left(\mathbf{1} + \mathrm{i}\frac{\tau}{2} \mathsf{H}_{\mathrm{ang}}^{(3)\ \ell m}\right)^{-1} \left(\mathbf{1} - \mathrm{i}\frac{\tau}{2} \mathsf{H}_{\mathrm{ang}}^{(3)\ \ell m}\right). \tag{53}$$

$\mathsf{R}_n^{\ell m}$ can be evaluated using Eq. (46):

$$\mathsf{H}_{\mathrm{ang}}^{(1,2)\,\ell m} = \begin{pmatrix} 0 & s_{n\ell m} \\ s^*_{n\ell m} & 0 \end{pmatrix} \tag{54}$$

with

$$s_{n\ell m} = -\frac{\mathrm{i}A(t)}{r_n} t_{\ell m} + \Big(r_n E(t) + V_{\mathrm{ee}}^1(r_n, t)\Big) c_{\ell m} \tag{55}$$

leads to

$$\mathsf{R}_n^{\ell m} = \frac{1}{1 + |w_{n\ell m}|^2} \begin{pmatrix} 1 - |w_{n\ell m}|^2 & -2w_{n\ell m} \\ 2w^*_{n\ell m} & 1 - |w_{n\ell m}|^2 \end{pmatrix}, \tag{56}$$

$$w_{n\ell m} = \mathrm{i}\frac{\tau}{2} s_{\ell m} = \frac{\tau}{2} \left(\frac{A(t)}{r_n} t_{\ell m} + \mathrm{i}\Big(r_n E(t) + V_{\mathrm{ee}}^1(r_n, t)\Big) c_{\ell m} \right). \tag{57}$$

For $\mathsf{Z}_n^{\ell m}$ one obtains

$$\mathsf{Z}_n^{\ell m} = \frac{1}{1 + |v_{n\ell m}|^2} \begin{pmatrix} 1 - |v_{n\ell m}|^2 & -2v_{n\ell m} \\ 2v^*_{n\ell m} & 1 - |v_{n\ell m}|^2 \end{pmatrix}, \tag{58}$$

$$v_{n\ell m} = \mathrm{i}\frac{\tau}{2} V_{\mathrm{ee}}^2(r_n)\, q_{\ell m}. \tag{59}$$

Let us now turn to the factor $\left[\mathsf{X}_+^{\ell m}\right]^{-1} \mathsf{X}_-^{\ell m}$ which involves $\ell, \ell+1$-sub blocks *and* a non-diagonality in r. We factor-out $\tilde{\mathsf{M}}_1^{-1}$ and write

$$\left[\mathsf{X}_+^{\ell m}\right]^{-1} \mathsf{X}_-^{\ell m} = \left[\mathsf{Y}_+^{\ell m}\right]^{-1} \mathsf{Y}_-^{\ell m}, \qquad \mathsf{Y}_\pm^{\ell m} = \mathbf{1}_\ell \otimes \tilde{\mathsf{M}}_1 \pm \frac{\tau}{2} A(t) \mathsf{L}_{\ell m} \otimes \tilde{\Delta}_1. \tag{60}$$

The matrices $\mathsf{Y}_{\pm}^{\ell m}$ are not tridiagonal but block-tridiagonal only:

$$\mathsf{Y}_{\pm}^{\ell m}=\left(\begin{array}{cc|cc|cc|cc|cc}
\frac{4+x}{6} & \pm y g_{\ell m} & \frac{1}{6} & \pm g_{\ell m} & & & & & & \\
\pm y g_{\ell m} & \frac{4+x}{6} & \pm g_{\ell m} & \frac{1}{6} & & & & & & \\ \hline
\frac{1}{6} & \mp g_{\ell m} & \frac{2}{3} & & \frac{1}{6} & \pm g_{\ell m} & & & & \\
\mp g_{\ell m} & \frac{1}{6} & & \frac{2}{3} & \pm g_{\ell m} & \frac{1}{6} & & & & \\ \hline
& & \frac{1}{6} & \mp g_{\ell m} & \frac{2}{3} & & \frac{1}{6} & \pm g_{\ell m} & & \\
& & \mp g_{\ell m} & \frac{1}{6} & & \frac{2}{3} & \pm g_{\ell m} & \frac{1}{6} & & \\ \hline
& & & & \cdot & & \cdot & & \cdot & \\
& & & & & \cdot & & \cdot & & \cdot \\ \hline
& & & & & & \frac{1}{6} & \mp g_{\ell m} & \frac{4+x}{6} & \mp y g_{\ell m} \\
& & & & & & \mp g_{\ell m} & \frac{1}{6} & \mp y g_{\ell m} & \frac{4+x}{6}
\end{array}\right)$$

with $g_{\ell m} = \tau A(t) c_{\ell m}/4h$. The rank of these matrices is $2N_r$. It operates on vectors of the form

$$\mathbf{\Gamma}_{\ell m}=\Big(\boxed{\Phi_{\ell m}(r_1,t),\Phi_{\ell+1,m}(r_1,t)},\boxed{\Phi_{\ell m}(r_2,t),\Phi_{\ell+1,m}(r_2,t)},\ldots\Big)^{\top}.$$

The matrices $\mathsf{Y}_{\pm}^{\ell m}$ can be transformed into a sum of two tridiagonal matrices $\overline{\mathsf{Y}}_{1\pm}^{\ell m}$ and $\overline{\mathsf{Y}}_{2\pm}^{\ell m}$, acting in two distinct vector spaces. Since

$$\mathsf{B}\mathsf{L}_{\ell m}\mathsf{B}^{\top}=\begin{pmatrix} c_{\ell m} & 0 \\ 0 & -c_{\ell m}\end{pmatrix},\qquad \mathsf{B}=\frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 1 \\ -1 & 1\end{pmatrix}$$

one obtains

$$\overline{\mathsf{Y}}_{\pm}^{\ell m}=(\mathsf{B}\otimes\mathbf{1}_r)\mathsf{Y}_{\pm}^{\ell m}(\mathsf{B}\otimes\mathbf{1}_r)^{\top}=\overline{\mathsf{Y}}_{1\pm}^{\ell m}+\overline{\mathsf{Y}}_{2\pm}^{\ell m} \tag{61}$$

with

$$\overline{\mathsf{Y}}_{1\pm}^{\ell m}=\left(\begin{array}{cc|cc|cc|cc|cc}
\frac{4+x}{6}\pm y g_{\ell m} & 0 & \frac{1}{6}\pm g_{\ell m} & 0 & & & & & & \\
0 & 0 & 0 & 0 & 0 & 0 & & & & \\ \hline
\frac{1}{6}\mp g_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6}\pm g_{\ell m} & 0 & & & & \\
0 & 0 & 0 & 0 & 0 & 0 & & & & \\ \hline
& & & & \ddots & \ddots & & & & \\ \hline
& & & & & & \frac{1}{6}\mp g_{\ell m} & 0 & \frac{4+x}{6}\mp y g_{\ell m} & 0 \\
& & & & & & 0 & 0 & 0 & 0
\end{array}\right),$$

$$\overline{\mathsf{Y}}_{2\pm}^{\ell m}=\left(\begin{array}{cc|cc|cc|cc|cc}
0 & 0 & 0 & 0 & & & & & & \\
0 & \frac{4+x}{6}\mp y g_{\ell m} & 0 & \frac{1}{6}\mp g_{\ell m} & & & & & & \\ \hline
0 & 0 & 0 & 0 & 0 & 0 & & & & \\
0 & \frac{1}{6}\pm g_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6}\mp g_{\ell m} & & & & \\ \hline
& & & & \ddots & \ddots & & & & \\ \hline
& & & & & & 0 & 0 & 0 & 0 \\
& & & & & & 0 & \frac{1}{6}\pm g_{\ell m} & 0 & \frac{4+x}{6}\pm y g_{\ell m}
\end{array}\right).$$

The vector $\mathbf{\Gamma}_{\ell m}$ transforms accordingly,

$$\overline{\mathbf{\Gamma}}_{\ell m} = (\mathsf{B} \otimes \mathbf{1}_r)\mathbf{\Gamma}_{\ell m} = \overline{\mathbf{\Gamma}}_{1\ell m} + \overline{\mathbf{\Gamma}}_{2\ell m} \tag{62}$$

where

$$\overline{\mathbf{\Gamma}}_{1\ell m} = \frac{1}{\sqrt{2}} \left(\begin{array}{c} \Phi^1_{\ell m} + \Phi^1_{\ell+1,m} \\ 0 \\ \hline \Phi^2_{\ell m} + \Phi^2_{\ell+1,m} \\ 0 \\ \hline \vdots \\ \hline \Phi^{N_r}_{\ell m} + \Phi^{N_r}_{\ell+1,m} \\ 0 \end{array} \right), \quad \overline{\mathbf{\Gamma}}_{2\ell m} = \frac{1}{\sqrt{2}} \left(\begin{array}{c} 0 \\ -\Phi^1_{\ell m} + \Phi^1_{\ell+1,m} \\ \hline 0 \\ -\Phi^2_{\ell m} + \Phi^2_{\ell+1,m} \\ \hline \vdots \\ \hline 0 \\ -\Phi^{N_r}_{\ell m} + \Phi^{N_r}_{\ell+1,m} \end{array} \right). \tag{63}$$

Now it is easy to see that

$$\overline{\mathsf{Y}}^{\ell m}_{\pm} \overline{\mathbf{\Gamma}}_{\ell m} = (\overline{\mathsf{Y}}^{\ell m}_{1\pm} + \overline{\mathsf{Y}}^{\ell m}_{2\pm})(\overline{\mathbf{\Gamma}}_{1\ell m} + \overline{\mathbf{\Gamma}}_{2\ell m}) = \overline{\mathsf{Y}}^{\ell m}_{1\pm}\overline{\mathbf{\Gamma}}_{1\ell m} + \overline{\mathsf{Y}}^{\ell m}_{2\pm}\overline{\mathbf{\Gamma}}_{2\ell m},$$

because the matrices $\overline{\mathsf{Y}}^{\ell m}_{1\pm}$, $\overline{\mathsf{Y}}^{\ell m}_{2\pm}$ operate in distinct spaces.

Finally, the factor $\mathsf{Q}_+^{-1}\mathsf{Q}_-$ in (50) may be written as

$$\mathsf{Q}_+^{-1}\mathsf{Q}_- = \mathsf{W}_+^{-1}\mathsf{W}_- \tag{64}$$

with

$$\mathsf{W}_\pm = \mathbf{1}_\ell \otimes \tilde{\mathsf{M}}_2 \pm \mathrm{i}\tau \mathbf{1}_\ell \otimes \left(\tilde{\Delta}_2 + \tilde{\mathsf{M}}_2(\mathsf{V}^\ell_{\text{eff}} + V^0_{\text{ee}} + p_{\ell m} V^2_{\text{ee}}) \right). \tag{65}$$

The matrices $\mathsf{W}_\pm$ are already diagonal in ℓ and tridiagonal in r and thus need no further treatment.

The short-time propagator for linear polarization (propagation mode `34`) finally reads

$$\begin{aligned} \mathsf{U}_{\text{CN}}(t+\Delta t, t) \quad = \quad & \prod_{\ell=L-3}^{0} \mathsf{Z}^{\ell m}_n \prod_{\ell=L-2}^{0} \left(\mathsf{R}^{\ell m}_n (\mathsf{B} \otimes \mathbf{1}_r)^\top \left[\overline{\mathsf{Y}}^{\ell m}_+\right]^{-1} \overline{\mathsf{Y}}^{\ell m}_- (\mathsf{B} \otimes \mathbf{1}_r) \right) \\ & \times \mathsf{W}_+^{-1}\mathsf{W}_- \\ & \times \prod_{\ell=0}^{L-2} \left((\mathsf{B} \otimes \mathbf{1}_r)^\top \left[\overline{\mathsf{Y}}^{\ell m}_+\right]^{-1} \overline{\mathsf{Y}}^{\ell m}_- (\mathsf{B} \otimes \mathbf{1}_r) \mathsf{R}^{\ell m}_n \right) \prod_{\ell=0}^{L-3} \mathsf{Z}^{\ell m}_n. \end{aligned} \tag{66}$$

3.3 Calculation of $r_<^L/r_>^{L+1}$-integrals

The integrals (31)–(33) are of the general form

$$F^L(r) = \int \mathrm{d}r' \, \frac{r_<^L}{r_>^{L+1}} \, f(r'), \qquad r_< = \min(r, r'), \quad r_> = \max(r, r'). \tag{67}$$

Integrals of the same kind will also appear further below when the KLI exchange potential is evaluated. Using the simple trapezoidal rule for numerical integration this translates to

$$F_j^L = \underline{F_j^L} + \overline{F_j^L}, \tag{68}$$

$$\underline{F_j^L} = \sum_{i=0}^{j-1} \Delta r f_i \frac{r_i^L}{r_j^{L+1}}, \qquad \overline{F_j^L} = \sum_{i=j}^{N_r-1} \Delta r f_i \frac{r_j^L}{r_i^{L+1}}. \tag{69}$$

The F_j^Ls can be calculated recursively,

$$\underline{F_{j+1}^L} = \frac{r_j^{L+1}}{r_{j+1}^{L+1}} \underline{F_j^L} + \Delta r f_j \frac{r_j^L}{r_{j+1}^{L+1}}, \qquad \overline{F_{j+1}^L} = \frac{r_{j+1}^L}{r_j^L} \overline{F_j^L} - \Delta r f_j \frac{r_{j+1}^L}{r_j^{L+1}}. \tag{70}$$

3.4 Exchange-correlation potential

There is a variety of approximations to the unknown exchange-correlation potential $V_{\mathrm{xc}\sigma}[n]$ in ground state DFT. In practice, the same functionals are often used in TDDFT, i.e., the stationary ground state KS orbitals are simply replaced by their time-dependent equivalents. In this paper we will discuss the implementation of the exchange potential proposed by Krieger, Li, and Iafrate (KLI) (cf. CPC-manuscript).

So-called "optimized effective potential" (OEP) methods take the exchange energy

$$\mathcal{E}_{\mathrm{x}} = -\frac{1}{2} \sum_{\sigma} \sum_{j,k=1}^{N_\sigma} \int \mathrm{d}^3 r \int \mathrm{d}^3 r' \, \frac{\Psi_{j\sigma}^*(\mathbf{r}) \Psi_{k\sigma}^*(\mathbf{r}') \Psi_{j\sigma}(\mathbf{r}') \Psi_{k\sigma}(\mathbf{r})}{|\mathbf{r} - \mathbf{r}'|} \tag{71}$$

exactly into account. The OEP method has been also extended to TDDFT. However, the OEP method yields a complicated integral equation for the exchange potential V_{x}. This OEP integral equation is impracticable for actual numerical implementations, especially in the time-dependent case. Krieger, Li, and Iafrate (KLI) proposed a method to solve the OEP integral equation approximately. The numerical KLI results yield highly accurate ionization potentials, for instance.

The KLI potential is given by

$$V_{\mathrm{x}\sigma}^{\mathrm{KLI}}(\mathbf{r}) = V_{\mathrm{x}\sigma}^{\mathrm{S}}(\mathbf{r}) + \tilde{V}_{\mathrm{x}\sigma}(\mathbf{r}) \tag{72}$$

where

$$\tilde{V}_{\mathrm{x}\sigma}(\mathbf{r}) = \sum_{i=1}^{N_\sigma - 1} \frac{|\Psi_{i\sigma}(\mathbf{r})|^2}{n_\sigma(\mathbf{r})} Q_{i\sigma}, \tag{73}$$

$$Q_{i\sigma} = \langle V_{\mathrm{x}\sigma}^{\mathrm{KLI}} \rangle_{i\sigma} - \langle u_{\mathrm{x}i\sigma} \rangle_{i\sigma}, \tag{74}$$

$$u_{\mathrm{x}i\sigma}(\mathbf{r}) = \frac{1}{\Psi_{i\sigma}^*(\mathbf{r})} \frac{\delta \mathcal{E}_{\mathrm{x}}}{\delta \Psi_{i\sigma}(\mathbf{r})} = -\sum_{j=1}^{N_\sigma} \frac{\Psi_{j\sigma}^*(\mathbf{r})}{\Psi_{i\sigma}^*(\mathbf{r})} \int \mathrm{d}^3 r' \, \frac{\Psi_{i\sigma}^*(\mathbf{r}') \Psi_{j\sigma}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \tag{75}$$

and

$$V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r}) = \sum_{i=1}^{N_\sigma} \frac{|\Psi_{i\sigma}(\mathbf{r})|^2}{n_\sigma(\mathbf{r})} \Re u_{\mathrm{x}i\sigma}(\mathbf{r}) \tag{76}$$

is the Slater potential. $\Re$ denotes the real part. $\langle X \rangle_{i\sigma}$ is the spatial average of an entity $X(\mathbf{r})$ weighted by the orbital spin density $|\Psi_{i\sigma}(\mathbf{r})|^2$,

$$\langle X \rangle_{i\sigma} = \int \mathrm{d}^3r \, |\Psi_{i\sigma}(\mathbf{r})|^2 X(\mathbf{r}).$$

As mentioned above, it is understood that in the time-dependent version of KLI all arguments $(\mathbf{r})$ are replaced by $(\mathbf{r}, t)$.

For calculating the ground state, $u_{\mathrm{x}i\sigma}$ can be chosen real, so that $\Re$ may be dropped in (76). The sum in (72) excludes the highest occupied orbital since it can be shown that

$$\langle V^{\mathrm{KLI}}_{\mathrm{x}\sigma} \rangle_{N_\sigma\sigma} = \langle u_{\mathrm{x}i\sigma} \rangle_{N_\sigma\sigma}. \tag{77}$$

The numbers $\langle V^{\mathrm{KLI}}_{\mathrm{x}\sigma} \rangle_{i\sigma}$ in (72) are calculated by solving the matrix equation

$$\sum_{i=1}^{N_\sigma - 1} (\delta_{ji} - M_{ji\sigma}) Q_{i\sigma} = \langle V^{\mathrm{S}}_{\mathrm{x}\sigma} \rangle_{j\sigma} - \langle u_{\mathrm{x}j\sigma} \rangle_{j\sigma} \tag{78}$$

with

$$M_{ji\sigma} = \int \mathrm{d}^3r \, \frac{|\Psi_{j\sigma}(\mathbf{r})|^2 |\Psi_{i\sigma}(\mathbf{r})|^2}{n_\sigma(\mathbf{r})} \tag{79}$$

for $Q_{i\sigma}$.

3.5 Calculation of $V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r})$ and $\tilde{V}_{\mathrm{x}\sigma}(\mathbf{r})$

Using (11), (23), and the Clebsch-Gordan coefficients $C^{c\gamma}_{a\alpha b\beta}$, the Slater potential may be written as

$$\begin{aligned} V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r}) \;&=\; -\frac{1}{\sqrt{4\pi} r^2 n_\sigma(\mathbf{r})} \sum_{i,j=1}^{N_\sigma} \sum_{L\tilde{L}\ell} \sum_{M\tilde{M}m} (-1)^M \frac{2\tilde{L}+1}{\sqrt{2\ell+1}} C^{\ell 0}_{L0\tilde{L}0} C^{\ell m}_{LM\tilde{L}\tilde{M}} \\ &\quad \times \tilde{\Lambda}^{\tilde{L}M^*}_{ji}(r) \int \mathrm{d}r' \, \frac{r_<^L}{r_>^{L+1}} \tilde{\Lambda}^{LM}_{ji}(r') Y^m_\ell(\Omega) \end{aligned} \tag{80}$$

where

$$\tilde{\Lambda}^{LM}_{ji}(r) = \sum_{\ell\ell'} \sqrt{\frac{2\ell+1}{2\ell'+1}} C^{\ell' 0}_{\ell 0 L 0} C^{\ell' m_j}_{\ell m_i L M} \Phi^*_{i\ell m_i}(r) \Phi_{j\ell' m_j}(r). \tag{81}$$

Note that only $M = m_j - m_i$, $\tilde{M} = -M$, and $m = 0$ contribute to the sums over M, $\tilde{M}$, and m so that (80) can be reduced to

$$\begin{aligned} V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r}) \;\; &= \;\; -\frac{1}{\sqrt{4\pi} r^2 n_\sigma(\mathbf{r})} \sum_{i,j=1}^{N_\sigma} \sum_{L\tilde{L}\ell} (-1)^{m_j - m_i} \frac{2\tilde{L}+1}{\sqrt{2\ell+1}} C^{\ell 0}_{L0\tilde{L}0} C^{\ell 0}_{Lm_j-m_i\tilde{L}m_i-m_j} \\ &\quad \times \tilde{\Lambda}^{\tilde{L}m_j-m_i\,*}_{ji}(r) \int \mathrm{d}r' \, \frac{r_<^L}{r_>^{L+1}} \tilde{\Lambda}^{Lm_j-m_i}_{ji}(r') Y^0_\ell(\Omega) \end{aligned} \tag{82}$$

with $\tilde{\Lambda}^L_{ji} := \tilde{\Lambda}^{Lm_j-m_i}_{ji}$. In passing we note that $\Lambda(r) = 2\sum_{i=1}^{N_\sigma} \tilde{\Lambda}^0_{ii}(r)$ since $C^{c\gamma}_{a\alpha 00} = \delta_{ac}\delta_{\alpha\gamma}$.

Expression (82) gives us a multipole expansion of $n_\sigma(\mathbf{r}) V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r})$. However, an expansion of $V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r})$ itself is needed in our numerical scheme. Hence, the factor $n_\sigma(\mathbf{r})^{-1}$ should, in principle, also be expanded in spherical harmonics, making the multipole expansion of $V^{\mathrm{S}}_{\mathrm{x}\sigma}(\mathbf{r})$ even more complicated. We thus, in this work, restrict ourselves to KLI in central field approximation (CFA). Note that the CFA is a prerequisite to obtain ground state KS orbitals of the form (12) at all.

In CFA, expression (82), upon neglecting all terms with $\ell > 0$ and replacing $n_\sigma(\mathbf{r})$ by $n_\sigma(r) = \Lambda(r)/(8\pi r^2)$ with $\Lambda(r)$ according (28) (remember assumption (B), Eq. (8)), simplifies to

$$V^{\mathrm{S}\,0}_{\mathrm{x}\sigma}(r) = -\frac{2}{\Lambda(r)} \sum_{i,j=1}^{N_\sigma} \sum_L \tilde{\Lambda}^{L\,*}_{ji}(r) \int \mathrm{d}r' \, \tilde{\Lambda}^L_{ji}(r') \frac{r_<^L}{r_>^{L+1}} \tag{83}$$

(note that $C^{00}_{a\alpha b\beta} = (-1)^{a-\alpha}\delta_{ab}\delta_{\alpha-\beta}/\sqrt{2a+1}$).

If one is just interested in the ground state Slater potential of closed-shell systems, (80) can be significantly simplified further. Making use of the form (12) for the ground state KS orbitals, rewriting sums over all orbitals like

$$\sum_{i=1}^{N_\sigma} \cdots = \sum_{i=1}^{N_{\mathrm{shells}}} \sum_{m_i=-\ell_i}^{\ell_i} \cdots \tag{84}$$

with i running over the different ℓ-shells now, and making use of the unitarity relation for Clebsch-Gordan coefficients[2] one arrives at

$$\begin{aligned} V^{\mathrm{S\,gs}}_{\mathrm{x}\sigma}(r) \;\; &= \;\; -\frac{2}{\Lambda(r)} \sum_{i,j=1}^{N_\sigma} \Phi_{i\sigma\ell_i}(r) \Phi^*_{j\sigma\ell_j}(r) \\ &\quad \times \sum_L \frac{\left[C^{\ell_j 0}_{\ell_i 0 L 0}\right]^2}{2\ell_j+1} \int \mathrm{d}r' \, \Phi^*_{i\sigma\ell_i}(r') \Phi_{j\sigma\ell_j}(r') \frac{r_<^L}{r_>^{L+1}} \end{aligned} \tag{85}$$

[2] When relations like $\sum_{\alpha\beta} C^{c\gamma}_{a\alpha b\beta} C^{c'\gamma'}_{a\alpha b\beta} = \delta_{cc'}\delta_{\gamma\gamma'}$ are used it is less error-prone to start with expressions like (80) where the sums over M, $\tilde{M}$, and m are still retained.

where we revoked (84) for both i and j, e.g., $(2\ell_i+1)\sum_{i=1}^{N_{\rm shells}} = \sum_{i=1}^{N_\sigma}$. The upper index 0 at $V_{\mathrm{x}\sigma}^{\mathrm{S}}$ (indicating the monopole term) has been dropped since closed-shell systems are exactly spherical. Note that the main steps that lead to (85) relied on the fact that the sums over m_i and n_j were complete (closed shells) and that the radial wave functions do not depend on m_i and m_j (spherical symmetry).

For $\tilde{V}_{\mathrm{x}\sigma}(\mathbf{r})$ one finds

$$\tilde{V}_{\mathrm{x}\sigma}(\mathbf{r}) = \frac{1}{\sqrt{4\pi}r^2 n_\sigma(\mathbf{r})} = \sum_{i=1}^{N_\sigma-1} Q_{i\sigma}\sum_L \sqrt{2L+1}\;\tilde{\Lambda}_{ii}^{L}(r)\,Y_L^0(\Omega), \tag{86}$$

$$\tilde{V}_{\mathrm{x}\sigma}^{0}(r) = \frac{2}{\Lambda(r)}\sum_{i=1}^{N_\sigma-1} Q_{i\sigma}\tilde{\Lambda}_{ii}^{0}(r), \tag{87}$$

and

$$\tilde{V}_{\mathrm{x}\sigma}^{\mathrm{gs}}(r) = \frac{2}{\Lambda(r)}\sum_{i=1}^{N_\sigma-1} Q_{i\sigma}|\Phi_{i\sigma\ell_i}(r)|^2 \tag{88}$$

with $Q_{i\sigma}$ given by (74).

3.6 Calculation of $\langle u_{\mathrm{x}j\sigma}\rangle_{j\sigma}$, $\langle V_{\mathrm{x}\sigma}^{\mathrm{S}}\rangle_{j\sigma}$, and $M_{ji\sigma}$

By performing a very similar calculation as in the previous section for $V_{\mathrm{x}\sigma}^{\mathrm{S}}(\mathbf{r})$ one obtains

$$\langle u_{\mathrm{x}j\sigma}\rangle_{j\sigma} = -\sum_{k=1}^{N_\sigma}\sum_L \int\!\mathrm{d}r\,\tilde{\Lambda}_{kj}^{L\,*}(r)\int\!\mathrm{d}r'\,\tilde{\Lambda}_{kj}^{L}(r')\frac{r_<^L}{r_>^{L+1}}, \tag{89}$$

and for the ground state

$$\begin{aligned}\langle u_{\mathrm{x}j\sigma}\rangle_{j\sigma}^{\mathrm{gs}} \;&=\; -\sum_{k=1}^{N_\sigma}\sum_L \frac{\left[C_{\ell_j 0 L 0}^{\ell_k 0}\right]^2}{2\ell_k+1}\int\!\mathrm{d}r\,\Phi_{j\sigma\ell_j}(r)\Phi_{k\sigma\ell_k}^{*}(r) \\ &\quad\times\int\!\mathrm{d}r'\,\Phi_{j\sigma\ell_j}^{*}(r')\Phi_{k\sigma\ell_k}(r')\frac{r_<^L}{r_>^{L+1}}.\end{aligned} \tag{90}$$

For $\langle V_{\mathrm{x}\sigma}^{\mathrm{S}}\rangle_{j\sigma}$

$$\langle V_{\mathrm{x}\sigma}^{\mathrm{S}}\rangle_{j\sigma} \;=\; -\int\!\mathrm{d}r\,\frac{2}{\Lambda(r)}\sum_{i,k=1}^{N_\sigma}\sum_{L\tilde{L}\ell}\tilde{\Lambda}_{jj}^{\ell\,*}(r)(2\ell+1)C_{L0\ell0}^{\tilde{L}0}C_{Lm_k-m_i\ell0}^{\tilde{L}m_k-m_i} \tag{91}$$

$$\times\tilde{\Lambda}_{ki}^{\tilde{L}\,*}(r)\int\!\mathrm{d}r'\,\frac{r_<^L}{r_>^{L+1}}\tilde{\Lambda}_{ki}^{L}(r') \tag{92}$$

and

$$\langle V_{x\sigma}^{\mathrm{S}}\rangle_{j\sigma}^{\mathrm{gs}} = -\int\!\mathrm{d}r\,\frac{2}{\Lambda(r)}|\Phi_{j\sigma\ell_j}(r)|^2\sum_{i,k=1}^{N_\sigma}\Phi_{i\sigma\ell_i}(r)\Phi_{k\sigma\ell_k}^*(r) \times\sum_L\frac{\left[C_{\ell_i0L0}^{\ell_k0}\right]^2}{2\ell_k+1}\int\!\mathrm{d}r'\,\Phi_{i\sigma\ell_i}^*(r')\Phi_{k\sigma\ell_k}(r')\frac{r_<^L}{r_>^{L+1}} \tag{93}$$

are obtained.

The matrix elements (79) are given by

$$M_{ji\sigma} = \int\!\mathrm{d}r\,\frac{2}{\Lambda(r)}\sum_L(2L+1)\,\tilde{\Lambda}_{jj}^{L\,*}(r)\,\tilde{\Lambda}_{ii}^{L\,*}(r) \tag{94}$$

and simplify in the case of closed-shell ground state systems to

$$M_{ji\sigma}^{\mathrm{gs}} = \tilde{M}_{ji\sigma}^{\mathrm{gs}}\sum_L(2L+1)\,C_{\ell_j0L0}^{\ell_j0}C_{\ell_jm_jL0}^{\ell_jm_j}C_{\ell_i0L0}^{\ell_i0}C_{\ell_im_iL0}^{\ell_im_i} \tag{95}$$

where

$$\tilde{M}_{ji\sigma}^{\mathrm{gs}} = \int\!\mathrm{d}r\,\frac{2}{\Lambda(r)}|\Phi_{j\sigma\ell_j}(r)|^2|\Phi_{i\sigma\ell_i}(r)|^2. \tag{96}$$

An alternative expression reads

$$M_{ji\sigma}^{\mathrm{gs}} = \tilde{M}_{ji\sigma}^{\mathrm{gs}}\sum_{LM}\frac{(2\ell_j+1)(2\ell_i+1)}{2L+1}\left[C_{\ell_j0\ell_i0}^{L0}\right]^2\left[C_{\ell_j-m_j\ell_im_i}^{LM}\right]^2. \tag{97}$$

Although expressions (95) and (97) depend on the m-quantum numbers m_j and m_i the right hand side of (78) must not depend on these in the case of closed-shell ground state systems. In fact, since $Q_{iN_\sigma}=0$, both $Q_{i\sigma}$ and $\tilde{M}_{ji\sigma}^{\mathrm{gs}}$ are independent of m_i,

$$\sum_{i=1}^{N_\sigma}M_{ji\sigma}^{\mathrm{gs}}Q_{i\sigma}^{\mathrm{gs}} = \sum_{i=1}^{N_{\mathrm{shells}}}\sum_{m_i}M_{ji\sigma}^{\mathrm{gs}}Q_{i\sigma}^{\mathrm{gs}}, \qquad \sum_{m_i}\sum_M\left[C_{\ell_j-m_j\ell_im_i}^{LM}\right]^2 = \frac{2L+1}{2\ell_j+1},$$

and $\sum_L(C_{\ell_j0\ell_i0}^{L0})^2=1$ one easily verifies that

$$\sum_{i=1}^{N_\sigma}M_{ji\sigma}^{\mathrm{gs}}Q_{i\sigma}^{\mathrm{gs}} = \sum_{i=1}^{N_\sigma}\tilde{M}_{ji\sigma}^{\mathrm{gs}}Q_{i\sigma}^{\mathrm{gs}}. \tag{98}$$

Thus, for calculating the ground state of closed-shell systems one can use $\tilde{M}_{ji\sigma}^{\mathrm{gs}}$ instead of the more complex $M_{ji\sigma}^{\mathrm{gs}}$ for solving the matrix equation (78).

4 Circular polarization: working in (r, ℓ, m)-space

In the case of a circularly polarized laser field the vector potential has (at least) two components. If the ionic potential has spherical symmetry and the dipole approximation can be applied one may choose the vector potential as

$$\mathbf{A}(t) = A_x(t)\mathbf{e}_x + A_y(t)\mathbf{e}_y. \tag{99}$$

The time-dependent Schrödinger equation then reads

$$\mathrm{i}\partial_t \Psi(\mathbf{r}, t) = \left(-\frac{1}{2}\nabla^2 + V(r) - \mathrm{i}A_x(t)\partial_x - \mathrm{i}A_y(t)\partial_y\right)\Psi(\mathbf{r}, t). \tag{100}$$

After an expansion in spherical harmonics one obtains

$$\begin{aligned}
\mathrm{i}\partial_t \Phi_{\ell m} &= \left(-\frac{1}{2}\frac{\partial^2}{\partial r^2} + V_\ell^{\mathrm{eff}}(r)\right)\Phi_{\ell m} \qquad (101)\\
&\quad -\frac{\mathrm{i}r}{2}\sum_{\ell' m'}\langle \ell m|[\exp(\mathrm{i}\varphi)\tilde{A}^* + \exp(-\mathrm{i}\varphi)\tilde{A}]\sin\vartheta|\ell' m'\rangle \partial_r \frac{1}{r}\Phi_{\ell' m'}\\
&\quad -\frac{\mathrm{i}}{2r}\sum_{\ell' m'}\langle \ell m|[\exp(\mathrm{i}\varphi)\tilde{A}^* + \exp(-\mathrm{i}\varphi)\tilde{A}]\cos\vartheta\partial_\vartheta|\ell' m'\rangle\Phi_{\ell' m'}\\
&\quad -\frac{\mathrm{i}}{2r}\sum_{\ell' m'}\langle \ell m|[\exp(\mathrm{i}\varphi)\tilde{A}^* - \exp(-\mathrm{i}\varphi)\tilde{A}]\frac{\mathrm{i}}{\sin\vartheta}\partial_\varphi|\ell' m'\rangle\Phi_{\ell' m'}\\
&= \left(-\frac{1}{2}\frac{\partial^2}{\partial r^2} + V_\ell^{\mathrm{eff}}(r)\right)\Phi_{\ell m}\\
&\quad -\frac{\mathrm{i}r}{2}\sum_{\ell' m'}\langle \ell m|[\exp(\mathrm{i}\varphi)\tilde{A}^* + \exp(-\mathrm{i}\varphi)\tilde{A}]\sin\vartheta|\ell' m'\rangle \partial_r \frac{1}{r}\Phi_{\ell' m'}\\
&\quad -\frac{\mathrm{i}}{2r}\sum_{\ell' m'}\langle \ell m|\tilde{A}^*\exp(\mathrm{i}\varphi)\left(\cos\vartheta\partial_\vartheta + \frac{\mathrm{i}}{\sin\vartheta}\partial_\varphi\right)|\ell' m'\rangle\Phi_{\ell' m'}\\
&\quad -\frac{\mathrm{i}}{2r}\sum_{\ell' m'}\langle \ell m|\tilde{A}\exp(-\mathrm{i}\varphi)\left(\cos\vartheta\partial_\vartheta - \frac{\mathrm{i}}{\sin\vartheta}\partial_\varphi\right)|\ell' m'\rangle\Phi_{\ell' m'},
\end{aligned}$$

where $|\ell m\rangle = |Y_\ell^m\rangle$ and $\tilde{A} = A_x + \mathrm{i}A_y$ have been introduced. If the ladder operators $\hat{\mathsf{L}}_\pm$ are defined as

$$\hat{\mathsf{L}}_\pm = -\frac{1}{\sqrt{2}}\exp(\pm\mathrm{i}\varphi)\left(\partial_\vartheta \pm \mathrm{i}\cot\vartheta\partial_\varphi\right) \tag{102}$$

they act on a spherical harmonic according

$$\hat{\mathsf{L}}_\pm|\ell m\rangle = \mp N_{\ell m}^\pm|\ell m \pm 1\rangle \tag{103}$$

where

$$N_{\ell m}^{\pm} = \sqrt{\frac{\ell(\ell+1) - m(m\pm 1)}{2}} = \sqrt{\frac{(\ell \mp m)(\ell \pm m + 1)}{2}}.$$

This may be used to rewrite the time-dependent Schrödinger equation as

$$\begin{aligned} \mathrm{i}\partial_t \Phi_{\ell m} &= \left(-\frac{1}{2}\frac{\partial^2}{\partial r^2} + V_\ell^{\mathrm{eff}}(r)\right)\Phi_{\ell m} \\ &\quad + \mathrm{i}\sqrt{\frac{2\pi}{3}} \sum_{\ell' m'} \Bigg\{ \tilde{A}^* \langle \ell m | 11 | \ell' m' \rangle \partial_r - \tilde{A} \langle \ell m | 1-1 | \ell' m' \rangle \partial_r \\ &\quad - \frac{\tilde{A}^*}{r} \langle \ell m | 11 | \ell' m' \rangle (1 + m') + \frac{\tilde{A}}{r} \langle \ell m | 1-1 | \ell' m' \rangle (1 - m') \\ &\quad - \frac{\tilde{A}^*}{r} \langle \ell m | 10 | \ell' m'+1 \rangle N_{\ell' m'}^{+} + \frac{\tilde{A}}{r} \langle \ell m | 10 | \ell' m'-1 \rangle N_{\ell' m'}^{-} \Bigg\} \Phi_{\ell' m'} \end{aligned} \tag{104}$$

with $\langle \ell m | LM | \ell' m' \rangle = \int \mathrm{d}\Omega \; Y_\ell^{m*} Y_L^M Y_{\ell'}^{m'}$. Three spherical harmonics integrated over the solid angle Ω may be expressed in terms of Clebsch-Gordan coefficients $C_{a\alpha b\beta}^{c\gamma}$,

$$\langle \ell m | LM | \ell' m' \rangle = \int \mathrm{d}\Omega \; Y_\ell^{m*} Y_L^M Y_{\ell'}^{m'} = \sqrt{\frac{(2L+1)(2\ell'+1)}{4\pi(2\ell+1)}} C_{\ell' 0 L 0}^{\ell 0} C_{\ell' m' LM}^{\ell m}.$$

The Clebsch-Gordon coefficients arising from the terms in (104) are quite simple and couple neighboring ℓs and ms only,

$$\begin{aligned} \langle \ell m | 11 | \ell' m' \rangle &= \sqrt{\frac{3}{4\pi}} \delta_{m,m'+1} \left(\delta_{\ell,\ell'+1} \sqrt{\frac{(\ell+m-1)(\ell+m)}{2(2\ell-1)(2\ell+1)}} \right. \\ &\qquad \left. - \delta_{\ell,\ell'-1} \sqrt{\frac{(\ell-m+1)(\ell-m+2)(\ell+1)}{(2\ell+1)(2\ell+2)(2\ell+3)}} \right), \\ \langle \ell m | 1-1 | \ell' m' \rangle &= \sqrt{\frac{3}{4\pi}} \delta_{m,m'-1} \left(\delta_{\ell,\ell'+1} \sqrt{\frac{(\ell-m-1)(\ell-m)}{2(2\ell-1)(2\ell+1)}} \right. \\ &\qquad \left. - \delta_{\ell,\ell'-1} \sqrt{\frac{(\ell+m+1)(\ell+m+2)(\ell+1)}{(2\ell+1)(2\ell+2)(2\ell+3)}} \right), \\ \langle \ell m | 10 | \ell' m'+1 \rangle &= \sqrt{\frac{3}{4\pi}} \delta_{m,m'+1} \left(\delta_{\ell,\ell'+1} \sqrt{\frac{(\ell+m)(\ell-m)}{(2\ell-1)(2\ell+1)}} \right. \\ &\qquad \left. + \delta_{\ell,\ell'-1} \sqrt{\frac{(\ell+m+1)(\ell-m+1)}{(2\ell+1)(2\ell+3)}} \right), \end{aligned}$$

$$\langle \ell m|10|\ell' m'-1\rangle \;=\; \sqrt{\frac{3}{4\pi}}\delta_{m,m'-1}\left(\delta_{\ell,\ell'+1}\sqrt{\frac{(\ell+m)(\ell-m)}{(2\ell-1)(2\ell+1)}}\right.$$
$$\left. +\delta_{\ell,\ell'-1}\sqrt{\frac{(\ell+m+1)(\ell-m+1)}{(2\ell+1)(2\ell+3)}}\right).$$

Using those results one arrives at

$$\begin{aligned}
\mathrm{i}\partial_t\Phi_{\ell m} = &\left(-\frac{1}{2}\frac{\partial^2}{\partial r^2}+V_\ell^{\mathrm{eff}}(r)\right)\Phi_{\ell m} \\
+\frac{\mathrm{i}}{2}\sum_{\ell' m'}\Bigg\{ \quad &\tilde{A}^*\delta_{m,m'+1}\delta_{\ell,\ell'+1}\sqrt{\frac{\ell+m}{(2\ell+1)(2\ell-1)}}\times \\
&\times\left[\left(\partial_r-\frac{m}{r}\right)\sqrt{\ell+m-1}\right. \\
&\left. -\frac{1}{r}\sqrt{(\ell-m)(\ell(\ell-1)-m(m-1))}\right] \\
&+\tilde{A}^*\delta_{m,m'+1}\delta_{\ell,\ell'-1}\sqrt{\frac{\ell-m+1}{(2\ell+1)(2\ell+3)}}\times \\
&\times\left[-\left(\partial_r-\frac{m}{r}\right)\sqrt{\ell-m+2}\right. \\
&\left. -\frac{1}{r}\sqrt{(\ell+m+1)((\ell+1)(\ell+2)-m(m-1))}\right] \\
&+\tilde{A}\delta_{m,m'-1}\delta_{\ell,\ell'+1}\sqrt{\frac{\ell-m}{(2\ell+1)(2\ell-1)}}\times \\
&\times\left[-\left(\partial_r+\frac{m}{r}\right)\sqrt{\ell-m-1}\right. \\
&\left. +\frac{1}{r}\sqrt{(\ell+m)(\ell(\ell-1)-m(m+1))}\right] \\
&+\tilde{A}\delta_{m,m'-1}\delta_{\ell,\ell'-1}\sqrt{\frac{\ell+m+1}{(2\ell+1)(2\ell+3)}}\times \\
&\times\left[\left(\partial_r+\frac{m}{r}\right)\sqrt{\ell+m+2}\right. \\
&\left. +\frac{1}{r}\sqrt{(\ell-m+1)((\ell+1)(\ell+2)-m(m+1))}\right] \quad \Bigg\}\Phi_{\ell' m'}
\end{aligned} \tag{105}$$

which may be written as

$$\mathrm{i}\partial_t\boldsymbol{\Phi} = \mathsf{H}\boldsymbol{\Phi}$$

where

$$\mathsf{H} = \mathsf{H}_{\text{at}} + \mathsf{H}_{\text{mix}} + \mathsf{H}_{\text{ang}}, \tag{106}$$

like in (36). H_{at} is diagonal in (ℓ, m)-space while H_{ang} is diagonal in r. The matrix components of H_{at}, H_{mix}, and H_{ang} are given by[3]

$$[\mathsf{H}_{\text{at}}]_{\ell m}^{\ell' m'} = \delta_{\ell,\ell'}\delta_{m,m'}\left(-\frac{1}{2}\frac{\partial^2}{\partial r^2} + V_\ell^{\text{eff}}(r)\right), \tag{107}$$

$$\begin{aligned}
[\mathsf{H}_{\text{ang}}]_{\ell m}^{\ell' m'} = \mathbf{1}_r \otimes \Big(&\mathcal{A}_{\ell m}\delta_{m,m'+1}\delta_{\ell,\ell'+1} + \mathcal{B}_{\ell m}\delta_{m,m'+1}\delta_{\ell,\ell'-1} \qquad (108)\\
&+\tilde{\mathcal{A}}_{\ell m}\delta_{m,m'-1}\delta_{\ell,\ell'+1} + \tilde{\mathcal{B}}_{\ell m}\delta_{m,m'-1}\delta_{\ell,\ell'-1}\Big),\\
[\mathsf{H}_{\text{mix}}]_{\ell m}^{\ell' m'} = \Big(&\mathcal{C}_{\ell m}\delta_{m,m'+1}\delta_{\ell,\ell'+1} + \mathcal{D}_{\ell m}\delta_{m,m'+1}\delta_{\ell,\ell'-1} \qquad (109)\\
&+\tilde{\mathcal{C}}_{\ell m}\delta_{m,m'-1}\delta_{\ell,\ell'+1} + \tilde{\mathcal{D}}_{\ell m}\delta_{m,m'-1}\delta_{\ell,\ell'-1}\Big)\ \partial_r
\end{aligned}$$

with

$$\begin{aligned}
\mathcal{A}_{\ell m} = \tilde{\mathcal{A}}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}^*}{2r}\sqrt{\frac{\ell+m}{(2\ell+1)(2\ell-1)}}\Big[&-m\sqrt{\ell+m-1}\\
&-\sqrt{(\ell-m)(\ell(\ell-1)-m(m-1))}\Big],
\end{aligned}$$

$$\begin{aligned}
\tilde{\mathcal{A}}_{\ell m} = \mathcal{A}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}}{2r}\sqrt{\frac{\ell-m}{(2\ell+1)(2\ell-1)}}\Big[&-m\sqrt{\ell-m-1}\\
&+\sqrt{(\ell+m)(\ell(\ell-1)-m(m+1))}\Big],
\end{aligned}$$

$$\begin{aligned}
\mathcal{B}_{\ell m} = \tilde{\mathcal{B}}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}^*}{2r}\sqrt{\frac{\ell-m+1}{(2\ell+1)(2\ell+3)}}\Big[&m\sqrt{\ell-m+2}\\
&-\sqrt{(\ell+m+1)((\ell+1)(\ell+2)-m(m-1))}\Big],
\end{aligned}$$

$$\begin{aligned}
\tilde{\mathcal{B}}_{\ell m} = \mathcal{B}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}}{2r}\sqrt{\frac{\ell+m+1}{(2\ell+1)(2\ell+3)}}\Big[&m\sqrt{\ell+m+2}\\
&+\sqrt{(\ell-m+1)((\ell+1)(\ell+2)-m(m+1))}\Big]
\end{aligned}$$

[3]The radius r is taken nondiscretized for the moment.

$$\mathcal{C}_{\ell m} = \tilde{\mathcal{C}}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}^*}{2}\sqrt{\frac{(\ell+m)(\ell+m-1)}{(2\ell+1)(2\ell-1)}},$$

$$\tilde{\mathcal{C}}_{\ell m} = \mathcal{C}^*_{\ell -m} = -\frac{\mathrm{i}\tilde{A}}{2}\sqrt{\frac{(\ell-m)(\ell-m-1)}{(2\ell+1)(2\ell-1)}},$$

$$\mathcal{D}_{\ell m} = \tilde{\mathcal{D}}^*_{\ell -m} = -\frac{\mathrm{i}\tilde{A}^*}{2}\sqrt{\frac{(\ell-m+1)(\ell-m+2)}{(2\ell+1)(2\ell+3)}},$$

$$\tilde{\mathcal{D}}_{\ell m} = \mathcal{D}^*_{\ell -m} = \frac{\mathrm{i}\tilde{A}}{2}\sqrt{\frac{(\ell+m+1)(\ell+m+2)}{(2\ell+1)(2\ell+3)}}.$$

The Hamiltonian H_{at} acts on each m-component of the wave function separately.

The wave function may be represented as a vector

$$\boldsymbol{\Phi} = \left(\boxed{\Phi_{00}}, \boxed{\Phi_{1-1}, \Phi_{10}, \Phi_{11}}, \boxed{\Phi_{2-2}, \Phi_{2-1}, \Phi_{20}, \Phi_{21}, \Phi_{22}}, \ldots, \boxed{\ldots, \Phi_{LL-1}, \Phi_{LL}}\right)^{\top}$$

where the ℓ-subblocks are indicated with boxes, and the discretized value of r is fixed.[4] In this representation $\mathsf{H}_{\mathrm{ang}}$ reads

[4]Since $\mathsf{H}_{\mathrm{ang}}$ is diagonal in r-space there is no need to indicate the value of r. $\mathsf{H}_{\mathrm{ang}}$ simply must be applied to *each* r-subblock.

$$
\mathsf{H}_{\mathrm{ang}} = \left(\begin{array}{c||c|ccc|ccccc|ccccccc}
 & 00 & 1\,{-1} & 10 & 11 & 2\,{-2} & 2\,{-1} & 20 & 21 & 22 & 3\,{-3} & 3\,{-2} & 3\,{-1} & 30 & 31 & 32 & 33 \\
\hline\hline
00 & & \mathcal{B}_{00} & & \tilde{\mathcal{B}}_{00} & & & & & & & & & & & & \\
\hline
1\,{-1} & \tilde{\mathcal{A}}_{1-1} & & & & \mathcal{B}_{1-1} & & \tilde{\mathcal{B}}_{1-1} & & & & & & & & & \\
10 & & & & & & \mathcal{B}_{10} & & \tilde{\mathcal{B}}_{10} & & & & & & & & \\
11 & \mathcal{A}_{11} & & & & & & \mathcal{B}_{11} & & \tilde{\mathcal{B}}_{11} & & & & & & & \\
\hline
2\,{-2} & & \tilde{\mathcal{A}}_{2-2} & & & & & & & & \mathcal{B}_{2-2} & & \tilde{\mathcal{B}}_{2-2} & & & & \\
2\,{-1} & & & \tilde{\mathcal{A}}_{2-1} & & & & & & & & \mathcal{B}_{2-1} & & \tilde{\mathcal{B}}_{2-1} & & & \\
20 & & \mathcal{A}_{20} & & \tilde{\mathcal{A}}_{20} & & & & & & & & \mathcal{B}_{20} & & \tilde{\mathcal{B}}_{20} & & \\
21 & & & \mathcal{A}_{21} & & & & & & & & & & \mathcal{B}_{21} & & \tilde{\mathcal{B}}_{21} & \\
22 & & & & \mathcal{A}_{22} & & & & & & & & & & \mathcal{B}_{22} & & \tilde{\mathcal{B}}_{22} \\
\hline
3\,{-3} & & & & & \tilde{\mathcal{A}}_{3-3} & & & & & & & & & & & \\
3\,{-2} & & & & & & \tilde{\mathcal{A}}_{3-2} & & & & & & & & & & \\
3\,{-1} & & & & & \mathcal{A}_{3-1} & & \tilde{\mathcal{A}}_{3-1} & & & & & & & & & \\
30 & & & & & & \mathcal{A}_{30} & & \tilde{\mathcal{A}}_{30} & & & & & & & & \\
31 & & & & & & & \mathcal{A}_{31} & & \tilde{\mathcal{A}}_{31} & & & & & & & \\
32 & & & & & & & & \mathcal{A}_{32} & & & & & & & & \\
33 & & & & & & & & & \mathcal{A}_{33} & & & & & & &
\end{array}\right) .
$$

Each matrix element left blank is zero. How the business continues for $\ell > 3$ is obvious.
The "mixing Hamiltonian" looks similarly, with $\mathcal{A}$ replaced by $\mathcal{C}$, and $\mathcal{B}$ with $\mathcal{D}$, but with an additional operator ∂_r.
Both $\mathsf{H}_{\mathrm{ang}}$ and $\mathsf{H}_{\mathrm{mix}}$ may be written as a sum over 2×2-matrices acting in (ℓ, m)-subspace,

$$
\mathsf{H}_{\mathrm{ang}} = \sum_{\ell=0}^{L-2} \sum_{m=-\ell}^{\ell} \left\{ \mathsf{H}_{\mathrm{ang}}^{\ell m} + \tilde{\mathsf{H}}_{\mathrm{ang}}^{\ell m} \right\}, \qquad \mathsf{H}_{\mathrm{mix}} = \sum_{\ell=0}^{L-2} \sum_{m=-\ell}^{\ell} \left\{ \mathsf{H}_{\mathrm{mix}}^{\ell m} + \tilde{\mathsf{H}}_{\mathrm{mix}}^{\ell m} \right\}, \tag{110}
$$

with

$$
\mathsf{H}_{\mathrm{ang}}^{\ell m} = \frac{\mathrm{i}|\tilde{A}|}{2r} \left(\begin{array}{c|cc} & \ell m & \ell+1, m-1 \\ \hline \ell m & 0 & \exp(-\mathrm{i}\phi) b_{\ell m} \\ \ell+1, m-1 & \exp(\mathrm{i}\phi) \tilde{a}_{\ell+1, m-1} & 0 \end{array} \right), \qquad \tilde{\mathsf{H}}_{\mathrm{ang}}^{\ell m} = \frac{\mathrm{i}|\tilde{A}|}{2r} \left(\begin{array}{c|cc} & \ell m & \ell+1, m+1 \\ \hline \ell m & 0 & \exp(\mathrm{i}\phi) \tilde{b}_{\ell m} \\ \ell+1, m+1 & \exp(-\mathrm{i}\phi) a_{\ell+1, m+1} & 0 \end{array} \right), \tag{111}
$$

$$\mathsf{H}^{\ell m}_{\mathrm{mix}} = -\frac{\mathrm{i}|\tilde{A}|}{2}\left(\begin{array}{c|cc} & \ell m & \ell+1, m-1 \\ \hline \ell m & 0 & \exp(-\mathrm{i}\phi) d_{\ell m} \\ \ell+1, m-1 & \exp(\mathrm{i}\phi)\tilde{c}_{\ell+1,m-1} & 0 \end{array}\right) \otimes \partial_r,$$

$$\tilde{\mathsf{H}}^{\ell m}_{\mathrm{mix}} = \frac{\mathrm{i}|\tilde{A}|}{2}\left(\begin{array}{c|cc} & \ell m & \ell+1, m+1 \\ \hline \ell m & 0 & \exp(\mathrm{i}\phi) \tilde{d}_{\ell m} \\ \ell+1, m+1 & \exp(-\mathrm{i}\phi) c_{\ell+1,m+1} & 0 \end{array}\right) \otimes \partial_r,$$

where

$$a_{\ell m} = \sqrt{\frac{\ell+m}{(2\ell+1)(2\ell-1)}}\left[-m\sqrt{\ell+m-1} - \sqrt{(\ell-m)(\ell(\ell-1)-m(m-1))}\right],$$

$$\tilde{a}_{\ell m} = \sqrt{\frac{\ell-m}{(2\ell+1)(2\ell-1)}}\left[-m\sqrt{\ell-m-1} + \sqrt{(\ell+m)(\ell(\ell-1)-m(m+1))}\right],$$

$$b_{\ell m} = \sqrt{\frac{\ell-m+1}{(2\ell+1)(2\ell+3)}}\left[m\sqrt{\ell-m+2} - \sqrt{(\ell+m+1)((\ell+1)(\ell+2)-m(m-1))}\right] = -\tilde{a}_{\ell+1,m-1},$$

$$\tilde{b}_{\ell m} = \sqrt{\frac{\ell+m+1}{(2\ell+1)(2\ell+3)}}\left[m\sqrt{\ell+m+2} + \sqrt{(\ell-m+1)((\ell+1)(\ell+2)-m(m+1))}\right] = -a_{\ell+1,m+1},$$

$$c_{\ell m} = \sqrt{\frac{(\ell+m)(\ell+m-1)}{(2\ell+1)(2\ell-1)}} = \tilde{c}_{\ell,-m} = \tilde{d}_{\ell-1,m-1},$$

$$\tilde{c}_{\ell m} = \sqrt{\frac{(\ell-m)(\ell-m-1)}{(2\ell+1)(2\ell-1)}} = c_{\ell,-m} = d_{\ell-1,m+1},$$

$$d_{\ell m} = \sqrt{\frac{(\ell-m+1)(\ell-m+2)}{(2\ell+1)(2\ell+3)}} = \tilde{d}_{\ell,-m},$$

$$\tilde{d}_{\ell m} = \sqrt{\frac{(\ell+m+1)(\ell+m+2)}{(2\ell+1)(2\ell+3)}} = d_{\ell,-m},$$

and the phase ϕ is defined through

$$\tilde{A} = |\tilde{A}|\exp(\mathrm{i}\phi).$$

$\mathsf{H}^{\ell m}_{\mathrm{ang}}$ and $\tilde{\mathsf{H}}^{\ell m}_{\mathrm{ang}}$ may be rewritten as

$$\mathsf{H}^{\ell m}_{\mathrm{ang}} = \frac{\mathrm{i}|\tilde{A}|}{2r} b_{\ell m} \underbrace{\left(\begin{array}{c|cc} & \ell m & \ell+1, m-1 \\ \hline \ell m & 0 & \exp(-\mathrm{i}\phi) \\ \ell+1, m-1 & -\exp(\mathrm{i}\phi) & 0 \end{array}\right)}_{\mathsf{P}^{\ell m}}, \tag{112}$$

$$\tilde{\mathsf{H}}^{\ell m}_{\text{ang}} = \frac{\mathrm{i}|\tilde{A}|}{2r}\tilde{b}_{\ell m} \underbrace{\left(\begin{array}{c|cc} & \ell m & \ell+1, m+1 \\ \hline \ell m & 0 & \exp(\mathrm{i}\phi) \\ \ell+1, m+1 & -\exp(-\mathrm{i}\phi) & 0 \end{array}\right)}_{\tilde{\mathsf{P}}^{\ell m}}. \tag{113}$$

$\mathsf{H}^{\ell m}_{\text{mix}}$ and $\tilde{\mathsf{H}}^{\ell m}_{\text{mix}}$ may be simplified as well:

$$\mathsf{H}^{\ell m}_{\text{mix}} = -\frac{\mathrm{i}|\tilde{A}|}{2}d_{\ell m} \underbrace{\left(\begin{array}{c|cc} & \ell m & \ell+1, m-1 \\ \hline \ell m & 0 & \exp(-\mathrm{i}\phi) \\ \ell+1, m-1 & \exp(\mathrm{i}\phi) & 0 \end{array}\right)}_{\mathsf{L}} \otimes \partial_r, \tag{114}$$

$$\tilde{\mathsf{H}}^{\ell m}_{\text{mix}} = \frac{\mathrm{i}|\tilde{A}|}{2}\tilde{d}_{\ell m} \underbrace{\left(\begin{array}{c|cc} & \ell m & \ell+1, m+1 \\ \hline \ell m & 0 & \exp(\mathrm{i}\phi) \\ \ell+1, m+1 & \exp(-\mathrm{i}\phi) & 0 \end{array}\right)}_{\tilde{\mathsf{L}}} \otimes \partial_r. \tag{115}$$

A Crank-Nicolson propagator U_{CN} which advances the wavefunction over $\Delta t = 2\tau$ may be chosen as follows:

$$\begin{aligned}
\mathsf{U}_{\text{CN}}(\tau) \;=\;& \prod_{\ell=L-2}^{0}\prod_{m=\ell}^{-\ell} \left(1+\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{ang}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{ang}}\right)\left(1+\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{mix}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{mix}}\right) \\
&\times \underbrace{\left(1+\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{ang}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{ang}}\right)}_{\tilde{\mathsf{R}}^{\ell m}} \underbrace{\left(1+\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{mix}}\right)}_{\tilde{\mathsf{X}}^{\ell m}_{+}}{}^{-1} \underbrace{\left(1-\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{mix}}\right)}_{\tilde{\mathsf{X}}^{\ell m}_{-}} \\
&\times \underbrace{(1+\mathrm{i}\tau\mathsf{H}_{\text{at}})}_{\mathsf{Q}_+}{}^{-1} \underbrace{(1-\mathrm{i}\tau\mathsf{H}_{\text{at}})}_{\mathsf{Q}_-} \\
&\times \prod_{\ell=0}^{L-2}\prod_{m=-\ell}^{\ell} \left(1+\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{mix}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{mix}}\right)\left(1+\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{ang}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\tilde{\mathsf{H}}^{\ell m}_{\text{ang}}\right) \\
&\times \underbrace{\left(1+\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{mix}}\right)}_{\mathsf{X}^{\ell m}_{+}}{}^{-1} \underbrace{\left(1-\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{mix}}\right)}_{\mathsf{X}^{\ell m}_{-}} \underbrace{\left(1+\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{ang}}\right)^{-1}\left(1-\mathrm{i}\frac{\tau}{2}\mathsf{H}^{\ell m}_{\text{ang}}\right)}_{\mathsf{R}^{\ell m}} \\
=\;& \prod_{\ell=L-2}^{0}\prod_{m=\ell}^{-\ell}\left(\mathsf{R}^{\ell m}\left[\mathsf{X}^{\ell m}_{+}\right]^{-1}\mathsf{X}^{\ell m}_{-}\tilde{\mathsf{R}}^{\ell m}\left[\tilde{\mathsf{X}}^{\ell m}_{+}\right]^{-1}\tilde{\mathsf{X}}^{\ell m}_{-}\right) \\
&\times \mathsf{Q}_+^{-1}\mathsf{Q}_- \prod_{\ell=0}^{L-2}\prod_{m=-\ell}^{\ell}\left(\left[\tilde{\mathsf{X}}^{\ell m}_{+}\right]^{-1}\tilde{\mathsf{X}}^{\ell m}_{-}\tilde{\mathsf{R}}^{\ell m}\left[\mathsf{X}^{\ell m}_{+}\right]^{-1}\mathsf{X}^{\ell m}_{-}\mathsf{R}^{\ell m}\right)
\end{aligned}$$

As in the simpler (r,ℓ)-case it is helpful to break the different factors of U_{CN}

down into digestible pieces:

$$\begin{aligned}
\left[\mathsf{X}_+^{\ell m}\right]^{-1}\mathsf{X}_-^{\ell m} &= \left(\mathbf{1}+\frac{\tau|\tilde{A}|d_{\ell m}}{4}\mathsf{L}\otimes\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1\right)^{-1}\left(\mathbf{1}-\frac{\tau|\tilde{A}|d_{\ell m}}{4}\mathsf{L}\otimes\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1\right)\\
&= \underbrace{\left(\mathbf{1}_{\ell m}\otimes\tilde{\mathsf{M}}_1+\frac{\tau|\tilde{A}|d_{\ell m}}{4}\mathsf{L}\otimes\tilde{\Delta}_1\right)}_{\mathsf{Y}_+^{\ell m}}^{-1}\underbrace{\left(\mathbf{1}_{\ell m}\otimes\tilde{\mathsf{M}}_1-\frac{\tau|\tilde{A}|d_{\ell m}}{4}\mathsf{L}\otimes\tilde{\Delta}_1\right)}_{\mathsf{Y}_-^{\ell m}},\\
\left[\tilde{\mathsf{X}}_+^{\ell m}\right]^{-1}\tilde{\mathsf{X}}_-^{\ell m} &= \left(\mathbf{1}-\frac{\tau|\tilde{A}|\tilde{d}_{\ell m}}{4}\tilde{\mathsf{L}}\otimes\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1\right)^{-1}\left(\mathbf{1}+\frac{\tau|\tilde{A}|\tilde{d}_{\ell m}}{4}\tilde{\mathsf{L}}\otimes\tilde{\mathsf{M}}_1^{-1}\tilde{\Delta}_1\right)\\
&= \underbrace{\left(\mathbf{1}_{\ell m}\otimes\tilde{\mathsf{M}}_1-\frac{\tau|\tilde{A}|\tilde{d}_{\ell m}}{4}\tilde{\mathsf{L}}\otimes\tilde{\Delta}_1\right)}_{\tilde{\mathsf{Y}}_+^{\ell m}}^{-1}\underbrace{\left(\mathbf{1}_{\ell m}\otimes\tilde{\mathsf{M}}_1+\frac{\tau|\tilde{A}|\tilde{d}_{\ell m}}{4}\tilde{\mathsf{L}}\otimes\tilde{\Delta}_1\right)}_{\tilde{\mathsf{Y}}_-^{\ell m}},\\
\mathsf{Q}_+^{-1}\mathsf{Q}_- &= \left(\mathbf{1}+\mathrm{i}\tau\mathbf{1}_{\ell m}\otimes(\tilde{\mathsf{M}}_2^{-1}\tilde{\Delta}_2+\mathsf{V}_{\text{eff}}^{\ell})\right)^{-1}\left(\mathbf{1}-\mathrm{i}\tau\mathbf{1}_{\ell m}\otimes(\tilde{\mathsf{M}}_2^{-1}\tilde{\Delta}_2+\mathsf{V}_{\text{eff}}^{\ell})\right)\\
&= \underbrace{\mathbf{1}_{\ell m}\otimes\left(\tilde{\mathsf{M}}_2+\mathrm{i}\tau(\tilde{\Delta}_2+\tilde{\mathsf{M}}_2\mathsf{V}_{\text{eff}}^{\ell})\right)}_{\mathsf{W}_+}^{-1}\underbrace{\mathbf{1}_{\ell m}\otimes\left(\tilde{\mathsf{M}}_2-\mathrm{i}\tau(\tilde{\Delta}_2+\tilde{\mathsf{M}}_2\mathsf{V}_{\text{eff}}^{\ell})\right)}_{\mathsf{W}_-}.
\end{aligned}$$

Let us first evaluate $\mathsf{R}^{\ell m}$ and $\tilde{\mathsf{R}}^{\ell m}$, respectively,

$$\begin{aligned}
\mathsf{R}^{\ell m} &= (1-\xi b_{\ell m}\mathsf{P}^{\ell m})^{-1}(1+\xi b_{\ell m}\mathsf{P}^{\ell m}), \qquad \xi=\frac{\tau|\tilde{A}|}{4r}\\
&= \frac{1}{1+\xi^2 b_{\ell m}^2}\begin{pmatrix} 1-\xi^2 b_{\ell m}^2 & 2\xi\exp(-\mathrm{i}\phi)b_{\ell m}\\ -2\xi\exp(\mathrm{i}\phi)b_{\ell m} & 1-\xi^2 b_{\ell m}^2\end{pmatrix},\\
\tilde{\mathsf{R}}^{\ell m} &= \frac{1}{1+\xi^2 \tilde{b}_{\ell m}^2}\begin{pmatrix} 1-\xi^2 \tilde{b}_{\ell m}^2 & 2\xi\exp(\mathrm{i}\phi)\tilde{b}_{\ell m}\\ -2\xi\exp(-\mathrm{i}\phi)\tilde{b}_{\ell m} & 1-\xi^2 \tilde{b}_{\ell m}^2\end{pmatrix}.
\end{aligned}$$

Next,

$$\begin{aligned}
\mathsf{Y}_\pm^{\ell m} &= \left(\tilde{\mathsf{M}}_1\pm\zeta d_{\ell m}\mathsf{L}\tilde{\Delta}_1\right), \qquad \zeta=\frac{\tau|\tilde{A}|}{4},\\
\tilde{\mathsf{Y}}_\pm^{\ell m} &= \left(\tilde{\mathsf{M}}_1\mp\zeta \tilde{d}_{\ell m}\tilde{\mathsf{L}}\tilde{\Delta}_1\right)
\end{aligned}$$

are tackled. Observing that with

$$\mathsf{B}=\frac{1}{\sqrt{2}}\begin{pmatrix}-\exp(\mathrm{i}\phi) & 1\\ \exp(\mathrm{i}\phi) & 1\end{pmatrix}, \qquad \mathsf{B}^{-1}=\mathsf{B}^\dagger=\frac{1}{\sqrt{2}}\begin{pmatrix}-\exp(-\mathrm{i}\phi) & \exp(-\mathrm{i}\phi)\\ 1 & 1\end{pmatrix}$$

and

$$\tilde{\mathsf{B}}=\frac{1}{\sqrt{2}}\begin{pmatrix}-\exp(-\mathrm{i}\phi) & 1\\ \exp(-\mathrm{i}\phi) & 1\end{pmatrix}, \qquad \tilde{\mathsf{B}}^{-1}=\tilde{\mathsf{B}}^\dagger=\frac{1}{\sqrt{2}}\begin{pmatrix}-\exp(\mathrm{i}\phi) & \exp(\mathrm{i}\phi)\\ 1 & 1\end{pmatrix}$$

one has

$$\mathsf{BLB}^\dagger = \tilde{\mathsf{B}}\tilde{\mathsf{L}}\tilde{\mathsf{B}}^\dagger = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}$$

it follows that

$$\begin{aligned}
\left[\mathsf{Y}_+^{\ell m}\right]^{-1}\mathsf{Y}_-^{\ell m} &= \mathsf{B}^\dagger\mathsf{B}\left[\mathsf{Y}_+^{\ell m}\right]^{-1}\mathsf{B}^\dagger\mathsf{B}\mathsf{Y}_-^{\ell m}\mathsf{B}^\dagger\mathsf{B} \\
&= \mathsf{B}^\dagger\left[\mathsf{B}\mathsf{Y}_+^{\ell m}\mathsf{B}^\dagger\right]^{-1}\mathsf{B}\mathsf{Y}_-^{\ell m}\mathsf{B}^\dagger\mathsf{B} \\
&= \mathsf{B}^\dagger\underbrace{\left[\tilde{\mathsf{M}}_1 + \zeta d_{\ell m}\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}\tilde{\Delta}_1\right]}_{\overline{\mathsf{Y}}_+^{\ell m}}^{-1}\underbrace{\left[\tilde{\mathsf{M}}_1 - \zeta d_{\ell m}\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}\tilde{\Delta}_1\right]}_{\overline{\mathsf{Y}}_-^{\ell m}}\mathsf{B}, \\
\left[\tilde{\mathsf{Y}}_+^{\ell m}\right]^{-1}\tilde{\mathsf{Y}}_-^{\ell m} &= \tilde{\mathsf{B}}^\dagger\underbrace{\left[\tilde{\mathsf{M}}_1 - \zeta\tilde{d}_{\ell m}\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}\tilde{\Delta}_1\right]}_{\overline{\tilde{\mathsf{Y}}}_+^{\ell m}}^{-1}\underbrace{\left[\tilde{\mathsf{M}}_1 + \zeta\tilde{d}_{\ell m}\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}\tilde{\Delta}_1\right]}_{\overline{\tilde{\mathsf{Y}}}_-^{\ell m}}\tilde{\mathsf{B}}
\end{aligned}$$

where the explicit indication of subspaces (i.e., the symbols $\mathbf{1}_{\ell m}$ and $\otimes$) was suppressed since it is clear in which spaces the matrices act. Like in the (r,ℓ)-case one may break the matrices $\overline{\mathsf{Y}}_\pm^{\ell m}$, $\overline{\tilde{\mathsf{Y}}}_\pm^{\ell m}$ down into a sum of two tridiagonal matrices acting in distinct vector spaces. Looking, for example, at one $(\ell m; \ell+1, m-1)$-subblock it is seen that

$$\begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}\tilde{\Delta}_1 = \frac{1}{2h}\left(\begin{array}{cc|cc|cc|cc}
-y & & -1 & & & & & \\
 & y & & 1 & & & & \\ \hline
1 & & & & -1 & & & \\
 & -1 & & & & 1 & & \\ \hline
 & & 1 & & & & -1 & \\
 & & & -1 & & & & 1 \\ \hline
 & & & & 1 & & y & \\
 & & & & & -1 & & -y
\end{array}\right)$$

where, just for illustration, the radial coordinate was discretized with four points only. Therefore, with $\zeta' = \zeta/2h$ one finds

$$\overline{\mathsf{Y}}_\pm^{\ell m} = \left(\begin{array}{cc|cc|cc|cc}
\frac{4+x}{6} \mp y\zeta' d_{\ell m} & & \frac{1}{6} \mp \zeta' d_{\ell m} & & & & & \\
 & \frac{4+x}{6} \pm y\zeta' d_{\ell m} & & \frac{1}{6} \pm \zeta' d_{\ell m} & & & & \\ \hline
\frac{1}{6} \pm \zeta' d_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \mp \zeta' d_{\ell m} & & & \\
 & \frac{1}{6} \mp \zeta' d_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \pm \zeta' d_{\ell m} & & \\ \hline
 & & \frac{1}{6} \pm \zeta' d_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \mp \zeta' d_{\ell m} & \\
 & & & \frac{1}{6} \mp \zeta' d_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \pm \zeta' d_{\ell m} \\ \hline
 & & & & \frac{1}{6} \pm \zeta' d_{\ell m} & & \frac{4+x}{6} \pm y\zeta' d_{\ell m} & \\
 & & & & & \frac{1}{6} \mp \zeta' d_{\ell m} & & \frac{4+x}{6} \mp y\zeta' d_{\ell m}
\end{array}\right),$$

$$
\overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm} = \left(\begin{array}{cc|cc|cc|cc}
\frac{4+x}{6} \pm y\zeta' \tilde{d}_{\ell m} & & \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & & & & & \\
& \frac{4+x}{6} \mp y\zeta' \tilde{d}_{\ell m} & & \frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} & & & & \\
\hline
\frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & & & \\
& \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} & & \\
\hline
& & \frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & \\
& & & \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & & \frac{2}{3} & & \frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} \\
\hline
& & & & \frac{1}{6} \mp \zeta' \tilde{d}_{\ell m} & & \frac{4+x}{6} \mp y\zeta' \tilde{d}_{\ell m} & \\
& & & & & \frac{1}{6} \pm \zeta' \tilde{d}_{\ell m} & & \frac{4+x}{6} \pm y\zeta' \tilde{d}_{\ell m}
\end{array}\right),
$$

where the result for $\overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm}$ is the same as the one for $\overline{\mathsf{Y}}^{\ell m}_{\pm}$ but with the replacements $\pm \to \mp$, $\mp \to \pm$, and $d_{\ell m} \to \tilde{d}_{\ell m}$. However, one must not forget that $\overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm}$ acts in the $(\ell m; \ell+1, m+1)$-subspace.

One may introduce the splitting

$$
\overline{\mathsf{Y}}^{\ell m}_{\pm} = \overline{\mathsf{Y}}^{\ell m}_{\pm 1} + \overline{\mathsf{Y}}^{\ell m}_{\pm 2}, \qquad \overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm} = \overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm 1} + \overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm 2}
$$

where

$$
\overline{\mathsf{Y}}^{\ell m}_{\pm 1} = \left(\begin{array}{cc|cc|cc|cc}
\frac{4+x}{6} \mp y\zeta' d_{\ell m} & 0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 & & & & \\
0 & 0 & 0 & 0 & & & & \\
\hline
\frac{1}{6} \pm \zeta' d_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 & & \\
0 & 0 & 0 & 0 & 0 & 0 & & \\
\hline
& & \frac{1}{6} \pm \zeta' d_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 \\
& & 0 & 0 & 0 & 0 & 0 & 0 \\
\hline
& & & & \frac{1}{6} \pm \zeta' d_{\ell m} & 0 & \frac{4+x}{6} \pm y\zeta' d_{\ell m} & 0 \\
& & & & 0 & 0 & 0 & 0
\end{array}\right),
$$

$$
\overline{\mathsf{Y}}^{\ell m}_{\pm 2} = \left(\begin{array}{cc|cc|cc|cc}
0 & 0 & 0 & 0 & & & & \\
0 & \frac{4+x}{6} \pm y\zeta' d_{\ell m} & 0 & \frac{1}{6} \pm \zeta' d_{\ell m} & & & & \\
\hline
0 & 0 & 0 & 0 & 0 & 0 & & \\
0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6} \pm \zeta' d_{\ell m} & & \\
\hline
& & 0 & 0 & 0 & 0 & 0 & 0 \\
& & 0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 & \frac{2}{3} & 0 & \frac{1}{6} \pm \zeta' d_{\ell m} \\
\hline
& & & & 0 & 0 & 0 & 0 \\
& & & & 0 & \frac{1}{6} \mp \zeta' d_{\ell m} & 0 & \frac{4+x}{6} \mp y\zeta' d_{\ell m}
\end{array}\right),
$$

and $\overline{\tilde{\mathsf{Y}}}^{\ell m}_{\pm 1,2}$ accordingly. Let us check how, for example, $\mathsf{B}^{\dagger} \left[\overline{\mathsf{Y}}^{\ell m}_{+}\right]^{-1} \overline{\mathsf{Y}}^{\ell m}_{-} \mathsf{B}$ acts on the wavefunction vector

$$
\mathbf{\Gamma}^{\ell m} = \left(\boxed{\Phi_{\ell m}(r_1), \Phi_{\ell+1,m-1}(r_1)}, \boxed{\Phi_{\ell m}(r_2), \Phi_{\ell+1,m-1}(r_2)}, \ldots\right)^{\top},
$$

corresponding to the (ℓm)-subblock under consideration. One finds

$$
\mathbf{\Lambda}^{\ell m} = \mathsf{B}^{\dagger} \left[\overline{\mathsf{Y}}^{\ell m}_{+}\right]^{-1} \overline{\mathsf{Y}}^{\ell m}_{-} \mathsf{B}\, \mathbf{\Gamma}^{\ell m} = \mathsf{B}^{\dagger} \left[\overline{\mathsf{Y}}^{\ell m}_{+}\right]^{-1} \overline{\mathsf{Y}}^{\ell m}_{-} (\overline{\mathbf{\Gamma}}^{\ell m}_{1} + \overline{\mathbf{\Gamma}}^{\ell m}_{2})
$$

where

$$
\overline{\mathbf{\Gamma}}^{\ell m}_{1} = \left(\boxed{-\exp(\mathrm{i}\phi)\Phi_{\ell m}(r_1) + \Phi_{\ell+1,m-1}(r_1), 0}, \boxed{-\exp(\mathrm{i}\phi)\Phi_{\ell m}(r_2) + \Phi_{\ell+1,m-1}(r_2), 0}, \ldots\right)^{\top},
$$

$$\overline{\mathbf{\Gamma}}_2^{\ell m} = \left(\boxed{0, \exp(\mathrm{i}\phi)\Phi_{\ell m}(r_1) + \Phi_{\ell+1,m-1}(r_1)}, \boxed{0, \exp(\mathrm{i}\phi)\Phi_{\ell m}(r_2) + \Phi_{\ell+1,m-1}(r_2)}, \ldots \right)^\top .$$

Since

$$\mathsf{B}^\dagger \left[\overline{\mathsf{Y}}_+^{\ell m}\right]^{-1} \left(\overline{\mathsf{Y}}_{-1}^{\ell m} + \overline{\mathsf{Y}}_{-2}^{\ell m}\right) \left(\overline{\mathbf{\Gamma}}_1^{\ell m} + \overline{\mathbf{\Gamma}}_2^{\ell m}\right)$$

one arrives with

$$\mathsf{B}\mathbf{\Lambda}^{\ell m} = \overline{\mathbf{\Lambda}}_1^{\ell m} + \overline{\mathbf{\Lambda}}_2^{\ell m}$$

at

$$\overline{\mathsf{Y}}_{+1}^{\ell m}\overline{\mathbf{\Lambda}}_1^{\ell m} + \overline{\mathsf{Y}}_{+2}^{\ell m}\overline{\mathbf{\Lambda}}_2^{\ell m} = \overline{\mathsf{Y}}_{-1}^{\ell m}\overline{\mathbf{\Gamma}}_1^{\ell m} + \overline{\mathsf{Y}}_{-2}^{\ell m}\overline{\mathbf{\Gamma}}_2^{\ell m} .$$

Because the vectors $\overline{\mathsf{Y}}_{+1}^{\ell m}\overline{\mathbf{\Lambda}}_1^{\ell m}$ and $\overline{\mathsf{Y}}_{-1}^{\ell m}\overline{\mathbf{\Gamma}}_1^{\ell m}$ lie in a vector space distinct to the one where $\overline{\mathsf{Y}}_{+2}^{\ell m}\overline{\mathbf{\Lambda}}_2^{\ell m}$ and $\overline{\mathsf{Y}}_{-2}^{\ell m}\overline{\mathbf{\Gamma}}_2^{\ell m}$ are members of, one can solve the two equations

$$\overline{\mathsf{Y}}_{+i}^{\ell m}\overline{\mathbf{\Lambda}}_i^{\ell m} = \overline{\mathsf{Y}}_{-i}^{\ell m}\overline{\mathbf{\Gamma}}_i^{\ell m}, \qquad i = 1, 2$$

separately for $\overline{\mathbf{\Lambda}}_i^{\ell m}$. In doing so one has to deal with tridiagonal matrices only. $\mathbf{\Lambda}^{\ell m}$ is obtained through $\mathbf{\Lambda}^{\ell m} = \mathsf{B}^\dagger\overline{\mathbf{\Lambda}}^{\ell m}$.

Finally, the short-time propagator for elliptic polarization (propagation mode 44) reads

$$\begin{aligned} \mathsf{U}_{\mathrm{CN}}(\tau) &= \prod_{\ell=L-2}^{0} \prod_{m=\ell}^{-\ell} \left(\mathsf{R}^{\ell m}\mathsf{B}^\dagger \left[\overline{\mathsf{Y}}_+^{\ell m}\right]^{-1} \overline{\mathsf{Y}}_-^{\ell m} \mathsf{B}\tilde{\mathsf{R}}^{\ell m}\tilde{\mathsf{B}}^\dagger \left[\overline{\tilde{\mathsf{Y}}}_+^{\ell m}\right]^{-1} \overline{\tilde{\mathsf{Y}}}_-^{\ell m} \tilde{\mathsf{B}} \right) \qquad (116) \\ &\quad \times \mathsf{W}_+^{-1}\mathsf{W}_- \prod_{\ell=0}^{L-2} \prod_{m=-\ell}^{\ell} \left(\tilde{\mathsf{B}}^\dagger \left[\overline{\tilde{\mathsf{Y}}}_+^{\ell m}\right]^{-1} \overline{\tilde{\mathsf{Y}}}_-^{\ell m} \tilde{\mathsf{B}}\tilde{\mathsf{R}}^{\ell m}\mathsf{B}^\dagger \left[\overline{\mathsf{Y}}_+^{\ell m}\right]^{-1} \overline{\mathsf{Y}}_-^{\ell m}\mathsf{B}\mathsf{R}^{\ell m} \right), \end{aligned}$$

where W_+, W_- are given in (65).