Unified view on linear response of interacting identical and distinguishable particles from multiconfigurational time-dependent Hartree methods

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Abstract

A unified view on linear response of interacting systems utilizing multiconfigurational time-dependent Hartree methods is presented. The cases of one-particle and two-particle response operators for identical particles and up to all-system response operators for distinguishable degrees-of-freedom are considered. The working equations for systems of identical bosons (LR-MCTDHB) and identical fermions (LR-MCTDHF), as well for systems of distinguishable particles (LR-MCTDH) are explicitly derived. These linear-response theories provide numerically-exact excitation energies and system’s properties, when numerical convergence is achieved in the calculations.

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I. INTRODUCTION

Excitation spectra of quantum systems are one of the widely used notions in our modern quantum world. They often allow one to explore the microscopic structure of various quantum objects, such as nuclei, atoms, molecules, and solids, and more recently, of ultracold quantum gases and Bose-Einstein condensates [1–8]. The excitation spectrum and excited states govern the spectroscopy and thermal properties of the quantum system of interest, and, of course, its quantum dynamics.

It is generally not possible to compute the excitation spectrum of the quantum object under interest analytically, except for a select few systems and models. One is opt then to resort to theoretical many-particle methods and to their numerical implementation. An often appealing and practical way to compute the excitation spectrum is linear response. Linear response is an in-principle exact approach, see, e.g., Refs. [9, 10], namely that one can obtain the exact excitation spectrum from the exact ground state, when weak perturbing fields are applied.

Notably the most famous linear-response theory is that for interacting identical fermions (electrons) whose wavefunction is described (approximated) by a single determinant; it is termed in different formulations time-dependent Hartree-Fock or random-phase approximation [11–13]. In many cases the ground state of electronic systems cannot be adequately described by a single determinant (configuration), and thus a multiconfigurational description becomes necessary – even more so for the linear-response computed excitations. In the context of atomic and molecular physics, linear response is often used in conjunction with self-consistent multiconfigurational and configuration-interaction ansätze, see, e.g., Refs. [10, 14, 15], and the review article Ref. [16] and references therein. We mention that for molecular systems and distinguishable degrees-of-freedom (vibrations), linear response for multiconfigurational wavefunctions has been put forward, see Refs. [17, 18].

For interacting identical bosons, which are traditionally assumed to be well described by a single permanent, mean-field wavefunction with all the bosons occupying the one and the same orbital, linear response also goes way back and is termed in different formulations Bogoliubov approximation or Bogoliubov–de Gennes equations [19–21]. With the advent of Bose-Einstein condensates made of ultracold quantum gases, this standard linear response has drawn much attention [20, 24].
It turns out that, unlike fermions, there are many distinct mean fields for bosons, because any number of bosons can occupy one, two, or many different orbitals. In other words, whereas a single configuration for fermions (i.e., a determinant, where each fermion ought to occupy a different orbital) is the only mean field for fermions, there are many distinct single configurations (i.e., permanents) for bosons, and hence many bosonic mean fields. For this plurality of bosonic mean fields and how to find among them the best (i.e., energetically lowest) mean field, see Refs. [25, 26]. In the context of linear response for bosonic systems, this has recently led to the linear-response theory of the best mean field [27], which unraveled excitations that cannot be found within the standard linear-response theory for bosons.

Before we proceed further, it should be mentioned for completeness that there is a huge literature on the computation and exploration of excitation spectra and excited states based on a variety of methods (either direct or/and in combination with linear response) such as: Configuration Interaction (also sometimes referred to as exact diagonalization); Coupled Cluster; Green’s Functions; Density Functional Theory; Quantum Monte Carlo; and more. These are not the direct subject of the present work.

In the present work we develop linear-response theories by linearizing numerically-exact evolution methods to solve the time-dependent many-particle Schrödinger equation. To be concrete, we derive and present a unified view of linear-response theories from the point of view of multiconfigurational time-dependent Hartree (MCTDH) propagation methods. The MCTDH method was invented in Refs. [28, 29] and is widely used for multi-dimensional dynamical systems consisting of distinguishable degrees-of-freedom, typically vibrations, see, e.g., Refs. [30–37], the reviews Refs. [38, 39] and references therein, and the software package Ref. [40]. MCTDH is considered at present the most efficient approach for accurate wavepacket propagation for distinguishable particles. MCTDH has been successfully extended to treat identical particles by accounting explicitly for the quantum statistics of the particles. MCTDH for fermions (MCTDHF) was derived in Refs. [41–45]; see, e.g., Refs. [46–55] for applications. MCTDH for bosons (MCTDHB) was derived in Refs. [56, 57]; for applications see, e.g., Refs. [58–63], and the software package Ref. [64]. We would like to mention that MCTDH itself has recently been applied with much success to a few-particle bosonic and fermionic systems, see, e.g., Refs. [65–70].

The equations of motion in MCTDH propagation methods originate directly from the time-dependent (Dirac-Frenkel) variational principle. As such, the coupled time-dependent
orbitals and expansion coefficients, and hence the many-particle wavefunction itself, are propagated in time in an optimal manner. Prescribing the linear response of the MCTDH propagation methods is hence a natural task. The MCTDH theories are very efficient, well-defined, commonly used, and formally exact, which also motivates us here to present the fundamental theory of their linearization.

Most recently, we have undertaken the respective task for bosons and presented the linear-response theory of MCTDHB \[71\]. It is a self-consistent multiconfigurational linear-response theory capable of computing exact many-body excitations of identical bosons, thus generalizing and extending the amply used, standard linear-response theory for Bose-Einstein condensates – Bogoliubov–de Gennes equations \[19–21\]. With increased capabilities to accurately measure excitation spectra of Bose-Einstein condensates in various trap potentials, see in this context Refs. \[72–75\], much is to be anticipated from the multiconfigurational linear-response theory of MCTDHB. The purpose of the present work is to build atop and expand on our recent findings for bosons. In what follows we derive the respective linear-response theories of the MCTDH propagation methods. This is done by linearizing the corresponding numerically-exact equations of motion for identical particles and distinguishable degrees-of-freedom, in a unified representation and manner.

The structure of the paper is as follows. In Sec. II we present a deductive exposition of the general feature that linear response atop the exact ground state provides the exact excitation spectra. We do so by treating the (already linear) time-dependent many-particle Schrödinger equation. In Sec. III we present the core linear-response theory for identical particles (bosons and fermions) based on MCTDHB and MCTDHF. We treat explicitly in the various subsections the different ingredients, up to the matrix form of the linear-response equations and utilizing its properties to solve the time-dependent identical-particle Schrödinger equation in linear response (Subsec. III F). We also utilize the projector operators in these MCTDH theories to arrive at an orthogonal response space, in which the orbitals’ and coefficients’ response amplitudes are explicitly orthogonal to the ground-state wavefunction. In Sec. IV we develop the linear-response theory for distinguishable degrees-of-freedom based on MCTDH, and discuss its structure and relation to the identical-particle theories. As a complementary result of the linear-response theories of Secs. III and IV we introduce into the MCTDH methods the notion of “fully-projected” equations of motion, which are obtained by adding to the famous orbitals’ differential condition \[28, 29\] a com-
II. LINEAR RESPONSE OF THE TIME-DEPENDENT (MANY-PARTICLE) SCHRÖDINGER EQUATION

We wish to show in this section, as a deductive and complementary preambles, that the linear response of the time-dependent Schrödinger equation with respect to perturbation of, typically, the ground state gives rise to the \textit{exact excitation spectrum} and corresponding eigenfunctions of the quantum system. Actually, no assumptions are made on the quantum system's Hamiltonian, except that the system is perturbed by a weak time-periodic field, hence – linear response. The derivation is inspired by \cite{22, 24, 27}. An additional purpose of this section is to introduce and clarify in a simpler, linear problem the role of projection operator(s) leading to fully orthogonal, or “fully projected” as we shall refer to it below, dynamics. This will become instrumental later on.

Let the time-independent Hamiltonian and Schrödinger equation read:

\begin{equation}
\hat{H}\Phi_k = E_k\Phi_k, \quad k = 0, 1, 2, \ldots,
\end{equation}

with the eigenvalues \(E_k\) and eigenvectors \(\Phi_k\) and for \(k = 0\) (typically) the ground state. We may write Eq. (1) equivalently as \(\hat{P}\Phi_k \hat{H}\Phi_k = 0, k = 0, 1, 2, \ldots\) with the projector \(\hat{P}\Phi_k = 1 - |\Phi_k\rangle\langle\Phi_k|\), also see below.

Now we wish to solve the time-dependent Schrödinger equation with the weak time-dependent \((\omega > 0)\) perturbation

\begin{equation}
\hat{H}(t)\Psi(t) = i\dot{\Psi}(t), \quad \dot{\Psi}(t) = \dot{\hat{H}} + \dot{f}e^{-i\omega t} + \dot{f}e^{+i\omega t}.
\end{equation}

We first perform a unitary transformation. By making the following assignment:

\begin{equation}
\Psi(t) \rightarrow \Psi(t)e^{-i\int_{t'}^{t'}(\Psi(t')|\hat{H}(t')|\Psi(t'))dt'}
\end{equation}

we obtain the “projected” time-dependent Schrödinger equation

\begin{equation}
\hat{P}\Psi \hat{H}(t)\Psi(t) = i\dot{\Psi}(t), \quad \hat{P} = 1 - |\Psi(t)\rangle\langle\Psi(t)|.
\end{equation}
The “projected” Schrödinger equation, Eq. (4), has the appealing property that the time-evolution is completely orthogonal in the sense that:

\[ i\langle \Psi(t)|\dot{\Psi}(t)\rangle = 0. \]

(5)

The differential condition Eq. (5) would relate later on to the familiar orbital differential condition introduced in Refs. [28, 29] and the below introduced coefficients’ differential condition in MCTDHB and MCTDHF, and in MCTDH theories.

In linear response we write for the solution of the time-dependent Schrödinger equation (4) [or, simply of Eq. (2) but then with the additional global phase \( e^{-iE_0t} \)] the ansatz:

\[ \Psi(t) \approx \Phi_0 + U e^{-i\omega t} + V^* e^{+i\omega t}. \]

(6)

Here and hereafter, the stationary state \( \Phi_0 \) is the zeroth-order approximation to \( \Psi(t) \) and the \( U \) and \( V^* \) are the first-order corrections or perturbations which are assumed to be small. Accordingly, in what follows the equations to be derived will be referred to as zeroth-order equations if they include only zeroth-order terms. The linear-response equations as well as other properties to be discussed will be referred to as first-order equations, since they are linear in the perturbations.

Making use of the differential condition Eq. (5), we immediately arrive at the orthogonality of the perturbed parts (for \( \omega > 0 \)) of the time-dependent wavefunction Eq. (6) with respect to the ground state:

\[ \langle \Phi_0 | U \rangle = 0, \quad \langle \Phi_0 | V^* \rangle = 0 \quad \iff \quad \hat{P}_{\Phi_0} U = U, \quad \hat{P}_{\Phi_0} V^* = V^*. \]

(7)

Inserting the ansatz Eq. (6) into Eq. (4), we obtain to zeroth order the stationary problem for the ground state, \( \hat{P}_{\Phi_0} \hat{H} \Phi_0 = 0 \). Furthermore, making use of Eq. (7) we arrive at the following result to first order:

\[ \hat{P}_{\Phi_0} (\hat{H} - E_0) (U e^{-i\omega t} + V^* e^{+i\omega t}) + \hat{P}_{\Phi_0} (\hat{f}^\dagger e^{-i\omega t} + \hat{f} e^{+i\omega t}) | \Phi_0 \rangle = \omega (U e^{-i\omega t} - V^* e^{+i\omega t}). \]

(8)

Next, equating same powers of \( e^{\pm i\omega t} \), adding a (redundant) projector in front of the perturbed parts of the wavefunction, \( U \) and \( V^* \), and collecting in a matrix form, we get as the final result for the equation of the perturbed time-dependent wavefunction:

\[
\begin{pmatrix}
\hat{P}_{\Phi_0} (\hat{H} - E_0) \hat{P}_{\Phi_0} & 0 \\
0 & -\hat{P}_{\Phi_0} (\hat{H}^* - E_0) \hat{P}_{\Phi_0}^*
\end{pmatrix}
- \omega
\begin{pmatrix}
1 & 0 \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
U \\
V
\end{pmatrix}
= \begin{pmatrix}
-\hat{P}_{\Phi_0} \hat{f}^\dagger | \Phi_0 \rangle \\
\hat{P}_{\Phi_0}^* \hat{f}^* | \Phi_0^* \rangle
\end{pmatrix}.
\]

(9)
To solve for Eq. (9) we first introduce the linear-response matrix $\mathcal{L}$:

$$
\mathcal{L} = \begin{pmatrix}
\hat{P}_{\Phi_0}(\hat{H} - E_0)\hat{P}_{\Phi_0} & 0 \\
0 & -\hat{P}_{\Phi_0}^*(\hat{H}^* - E_0)\hat{P}_{\Phi_0}^*
\end{pmatrix}
$$

and solve the linear-response eigenvalue system:

$$
\mathcal{L} \begin{pmatrix} U_k \\ V_k \end{pmatrix} = \omega_k \begin{pmatrix} U_k \\ V_k \end{pmatrix}
$$

associated with the Schrödinger equation. It is easily diagonalized and has two “branches”:

$$
\begin{pmatrix} U_k \\ V_k \end{pmatrix} = \begin{pmatrix} \Phi_k \\ 0 \end{pmatrix}, \quad \omega_k = E_k - E_0, \quad k = 0, 1, 2, \ldots,
$$

$$
\begin{pmatrix} U_{-k} \\ V_{-k} \end{pmatrix} = \begin{pmatrix} 0 \\ \Phi_k^* \end{pmatrix}, \quad -\omega_k, \quad k = 0, 1, 2, \ldots.
$$

Thus, it is readily seen that the linear-response of the Schrödinger equation (2) provides the exact excitation energies and corresponding excited-state eigenfunctions with respect to $\Phi_0$, which is usually the ground state. The so-called zero-mode excitations, which equal nothing but the ground-state energy of the system, separate the positive [$w_k > 0$; upper “branch” of Eq. (12)] and negative [$w_k < 0$; lower “branch” of Eq. (12)] parts of the spectrum.

Having solved the linear-response eigenvalue system, Eq. (11), we can now express the solution of Eq. (9) in terms of the excitation energies $\{\omega_k\}$ and eigenvectors.

To this end we expand the perturbed wavefunction:

$$
\begin{pmatrix} U \\ V \end{pmatrix} = \sum_k c_k \begin{pmatrix} U_k \\ V_k \end{pmatrix} = \sum_{k>0} \left\{ c_k \begin{pmatrix} \Phi_k \\ 0 \end{pmatrix} + c_{-k} \begin{pmatrix} 0 \\ \Phi_k^* \end{pmatrix} \right\}.
$$

The zero-mode, two $k = 0$ excitations do not have to be included in the expansion Eq. (13), because they give no contribution in view of the orthogonality relations in Eq. (7). The expansion coefficients thus read:

$$
c_k = \frac{\langle \Phi_k | \hat{f}^\dagger | \Phi_0 \rangle}{w - w_k}, \quad k = 1, 2, \ldots,
$$

$$
c_{-k} = -\frac{\langle \Phi_k^* | \hat{f}^* | \Phi_0^* \rangle}{w + w_k}, \quad k = 1, 2, \ldots,
$$

which completes the solution of the Schrödinger equation within linear response.
A final remark concerning hermiticity and completeness is in place. The linear-response of the (already linear) Schrödinger equation gives rise to the eigenvalue system Eq. (11) with an hermitian linear-response matrix $L$, Eq. (10) (for other characteristics of $L$ see above). Thus, the usual properties of normalization, orthogonality, and completeness of its eigenfunctions \[ \{ \left( \begin{array}{c} U_k \\ V_k \end{array} \right) \} \] simply hold. From them, the resolution of the identity:

\[
1 = \left( \begin{array}{c} \Phi_0 \\ 0 \end{array} \right) \left( \langle \Phi_0 | 0 \rangle \right) + \left( \begin{array}{c} 0 \\ \Phi_0^* \end{array} \right) \left( 0 \langle \Phi_0^* | 0 \rangle \right) + \sum_{k>0} \left( \begin{array}{c} \Phi_k \\ 0 \end{array} \right) \left( \langle \Phi_k | 0 \rangle \right) + \left( \begin{array}{c} 0 \\ \Phi_k^* \end{array} \right) \left( 0 \langle \Phi_k^* | 0 \rangle \right)
\]

(15)

and spectral resolution of the linear-response matrix:

\[
L = \sum_{k>0} \omega_k \left\{ \left( \begin{array}{c} \Phi_k \\ 0 \end{array} \right) \left( \langle \Phi_k | 0 \rangle \right) - \left( \begin{array}{c} 0 \\ \Phi_k^* \end{array} \right) \left( 0 \langle \Phi_k^* | 0 \rangle \right) \right\}
\]

(16)

follow. In the forthcoming sections, we discuss extensively the linear response of many-particle systems described by multiconfigurational time-dependent Hartree methods, which of course consist of nonlinear equations. The resulting response matrices, as in, e.g., Refs. [22, 24, 27], will no longer be hermitian. Hence, analogous resolutions like Eqs. (15) and (16) would be assumed to hold and then constructed.

III. LINEAR RESPONSE IN THE MULTICONFIGURATIONAL TIME-DEPENDENT HARTREE FRAMEWORK FOR IDENTICAL PARTICLES

This section deals with systems of interacting bosons or fermions and – starting from the propagation theories MCTDHB and MCTDHF – it derives their respective linear-response theories, which we denote by LR-MCTDHB and LR-MCTDHF, in a unified framework.

A. Basic and new ingredients

Consider a quantum object made of $N$ identical interacting particles, bosons or fermions. Our starting point is the MCTDHX (X=B,F) equations of motion, see Refs. [41–44, 56, 57], which can be represented in a unified manner [45]. The MCTDHX wavefunction is written as $|\Psi(t)\rangle = \sum_{\vec{n}} C_{\vec{n}} |\vec{n}; t\rangle$, where $|\vec{n}; t\rangle = \frac{1}{\sqrt{n_1! \cdots n_M!}} [\hat{c}_1(t)^\dagger]^{n_1} \cdots [\hat{c}_M(t)^\dagger]^{n_M} |\text{vac}\rangle$ are time-dependent configurations – either permanents for bosons or determinants for fermions. $\vec{n} =$
\((n_1, \ldots, n_M)\) is the vector of occupations and \(|\text{vac}\rangle\) the vacuum state. The creation \(\{\hat{c}_k^\dagger(t)\}\) and respective annihilation operators \(\{\hat{c}_k(t)\}\) are associated with a set of \(M\) time-dependent orthonormal orbitals \(\{\phi_k(\mathbf{r}, t)\}\). The vector of time-dependent coefficients \(\mathbf{C}(t) = \{C_n\}\) runs over all time-dependent configurations generated by distributing the \(N\) identical particles over the \(M\) orbitals. The number of such configurations is denoted hereafter by \(N_{\text{conf}}\).

The MCTDHX equations of motion are given by:

\[
\hat{P} \sum_{q=1}^{M} [\rho_{kq}\hat{\mathbf{p}} + \sum_{s,l=1}^{M} \rho_{kslq}\hat{W}_{sl}] |\phi_q\rangle = i \sum_{q=1}^{M} \rho_{kq} |\dot{\phi}_q\rangle, \quad k = 1, \ldots, M,
\]

\[
\mathbf{H}(t)\mathbf{C}(t) = i \frac{\partial \mathbf{C}(t)}{\partial t} \iff \mathbf{C}\hat{\mathbf{H}}(t) = i \dot{\mathbf{C}}(t), \quad H_{\vec{n}\vec{n}'}(t) = \langle \vec{n}; t | \hat{H} | \vec{n}'; t \rangle, \quad (17)
\]

with the local (direct) potentials:

\[
\hat{W}_{kq} = \int d\mathbf{r}' \phi_k^*(\mathbf{r}') \hat{W}(\mathbf{r} - \mathbf{r}') \phi_q(\mathbf{r}').
\]

Both reduced density matrices \([76, 78]\), which are highly helpful in MCTDHX theories \([79]\), and mapping of configurations and the operation of operators upon them \([80, 81]\) are used to shorten and simplify the notation. Occasionally we will use a double or mixed notation, especially for the equations of motion of the coefficients, when instructive. The first notation implies, throughout this work: \(\rho_{kq} = \langle \Psi | \hat{\rho}_{kq} | \Psi \rangle\) and \(\rho_{kslq} = \langle \Psi | \hat{\rho}_{kslq} | \Psi \rangle\), where \(\hat{\rho}_{kq} = \hat{c}_k^\dagger \hat{c}_q\) and \(\hat{\rho}_{kslq} = \hat{c}_k^\dagger \hat{c}_s^\dagger \hat{c}_l \hat{c}_q\) are the density operators. The second notation means, throughout this work, as follows. Consider a generic, second-quantized operator \(\hat{O}\) acting on the MCTDHX wavefunction. The operation readdresses the configurations, multiples them by respective matrix elements and numerical factors, and thereby changes the vector of coefficients \([80, 81]\): \(\hat{O} \sum_{\vec{n}} C_{\vec{n}} |\vec{n}; t \rangle \equiv \sum_{\vec{n}} C_{\vec{n}}^{\hat{O}} |\vec{n}; t \rangle\). The second summation means that the vector of changed time-dependent coefficients \(\mathbf{C}^{\hat{O}}(t) = \{C_{\vec{n}}^{\hat{O}}\}\) runs over the same time-dependent configurations generated by distributing the \(N\) identical particles over \(M\) orbitals. Below we treat the operation of one-body and two-body operators \(\hat{O}\). When needed and for the uniformity of the presentation, we exploit this notation also for the multiplication by a constant. For simplicity, we denote in Eqs. \((17)\) and \((18)\) the interaction \(\hat{W}(\mathbf{r} - \mathbf{r}')\) such that it depends, as is common, on the distance between the (identical) particles. The more general case of interaction between identical particles which is merely symmetric in the coordinates of the particles is implicitly included. Spin degrees-of-freedom are suppressed and implicit summation on them is assumed throughout this work and to be unambiguously performed.
The differential condition is satisfied by Eq. (17) and given by:

\[ i\langle \phi_k | \dot{\phi}_q \rangle = 0, \quad k, q = 1, \ldots, M. \]  

(19)

This guarantees the orthonormality of the orbitals at all times:

\[ \langle \phi_k | \phi_q \rangle = \delta_{kq}, \quad k, q = 1, \ldots, M. \]  

(20)

The projector is given by:

\[ \hat{P} = 1 - \sum_{j'=1}^{M} |\phi_{j'}\rangle \langle \phi_{j'}|. \]  

(21)

Finally, the time evolution of the coefficients is, of course, unitary which guarantees their normalization:

\[ C^\dagger C = 1. \]  

(22)

Except for a unified and practical way of writing in terms of reduced density matrices, equations of motion (17) provide the standard representation of MCTDHX theories. They describe the evolution of the orbitals in their orthogonal space [in view of the differential condition Eq. (19)] and the unitary evolution of the expansion coefficients. It turns out that with a single and simple unitary transformation one can achieve orthogonal propagation for the coefficients’ time evolution as well. This result stands for itself and will also prove to be instrumental for linear response. Explicitly, with the assignment of a joint time-dependent phase to all coefficients:

\[ C \rightarrow C e^{-i \int dt' C^\dagger(t')H(t')C(t')}, \]  

(23)

the “fully-projected” equations of motion of MCTDHX take on the novel form:

\[ \hat{P} \sum_{q=1}^{M} [\rho_{kq} \hat{h} + \sum_{s,l=1}^{M} \rho_{kstl} \hat{W}_{sl}] |\phi_q\rangle = i \sum_{q=1}^{M} \rho_{kq} |\dot{\phi}_q\rangle, \quad k = 1, \ldots, M, \]  

(24)

\[ \hat{P} C H(t) C(t) = i \frac{\partial C(t)}{\partial t} \iff \hat{P} C \hat{H}(t) = i \dot{C}(t) \iff C \hat{H} - C^\dagger C^\beta(t) = i \dot{C}(t), \]  

with the coefficients’ projector operator given by:

\[ \hat{P}_C = 1 - CC^\dagger. \]  

(25)

Now, also the coefficients’ part satisfies a differential condition:

\[ i C^\dagger \dot{C} = 0. \]  

(26)
Consequently and appealingly, by combining Eq. (19) for the orbitals and Eq. (26) for the expansion coefficients it is seen that the MCTDHX wavefunction $|\Psi(t)\rangle = \sum_{\vec{n}} C_{\vec{n}} |\vec{n}; t\rangle$ evolves in a completely orthogonal manner, namely $i \langle \Psi(t)|\dot{\Psi}(t)\rangle = 0$. This is just as the time-dependent wavefunction of the “projected” Schrödinger equation does, see Eq. (5).

We will also be needing the static (time-independent) theories of MCTDHX – MCHX (X=B,F), see Refs. [79, 82, 83]. This is because, as mentioned above, the linear response is to be performed around a static solution of the many-particle system, typically the ground state. The MCHX equations are given, e.g., by using imaginary-time propagation on MCTDHX, by:

$$\hat{P} \sum_{q=1}^{M} [\rho_{kj} \hat{h} + \sum_{s,l=1}^{M} \rho_{kslq} \hat{W}_{sl}] |\phi_q\rangle = 0, \quad k = 1, \ldots, M, \quad \Leftrightarrow \quad (27)$$

$$\sum_{q=1}^{M} [\rho_{kj} \hat{h} - \mu_{kq} + \sum_{s,l=1}^{M} \rho_{kslq} \hat{W}_{sl}] |\phi_q\rangle = 0, \quad k = 1, \ldots, M,$$

$$\mu_{kq} = \sum_{j=1}^{M} \langle \phi_q | [\rho_{kj} \hat{h} + \sum_{s,l=1}^{M} \rho_{kslj} \hat{W}_{sl}] |\phi_j\rangle, \quad k, q = 1, \ldots, M,$$

$$\mathbf{P}_C \mathbf{H} \mathbf{C} = \mathbf{0} \Leftrightarrow \mathbf{H} \mathbf{C} = \varepsilon \mathbf{C} \Leftrightarrow \mathbf{C}^{\hat{H}-\varepsilon \mathbf{C}} = 0 \Leftrightarrow \mathbf{C}^{\hat{H}-\varepsilon} = \mathbf{0}, \quad \mathbf{H}_{\vec{n}\vec{n}'} = \langle \vec{n}|\hat{h}|\vec{n}'\rangle.$$  

Note that in MCHX the matrix of Lagrange multipliers $\{\mu_{kq}\}$ is hermitian and can be diagonalized, namely

$$\mu_{kq}, \quad k, q = 1, \ldots, M \quad \longrightarrow \quad \mu_k, \quad k = 1, \ldots, M. \quad (28)$$

We also recall that the eigenenergy $\varepsilon$ is, in fact, a Lagrange multiplier that ensures (redundantly – in the time-dependent case) the vector of coefficients $\mathbf{C}$ to be normalized.

In the linear-response derivation that follows and whenever unambiguous, we denote the time-dependent and time-independent quantities by the same symbols, to avoid cumbersome notation.

**B. Perturbation and variation: Orthogonality**

We derive the linear-response theory from MCTDHX using a small perturbation around the MCHX solution, typically the ground state. Thus, we have the following ansatz for the
perturbing fields and the perturbed wavefunction:

\[ \phi_k(r, t) \approx \phi_k(r) + \delta \phi_k(r, t), \quad k = 1, \ldots, M, \]
\[ \delta \phi_k(r, t) = u_k(r)e^{-i\omega t} + v_k^*(r)e^{i\omega t}, \quad k = 1, \ldots, M, \]
\[ C(t) \approx [C + \delta C(t)] \quad \iff \quad C(t) \approx e^{-i\epsilon t}[C + \delta C(t)] \quad \text{(without coefficients' projector)}, \]
\[ \delta C(t) = Cu e^{-i\omega t} + C^*_v e^{i\omega t}, \]
\[ \delta h(r, t) = \hat{f}^\dagger(r)e^{-i\omega t} + \hat{f}(r)e^{i\omega t}, \]
\[ \delta W(r - r', t) = \hat{g}^\dagger(r - r')e^{-i\omega t} + \hat{g}(r - r')e^{i\omega t}. \] (29)

Here, \( \{\delta \phi_k(r, t)\} \) and \( \delta C(t) = \{\delta C_n(t)\} \) are the perturbed parts of the orbitals and coefficients, respectively, and comprised of \( u \) and \( v \) parts. The operators \( \hat{f}(r) \) and \( \hat{g}(r - r') \) generate one-body and two-body perturbations. We consider a time-dependent perturbation hence \( \omega > 0 \). The Hamiltonian including the perturbation is hermitian.

As mentioned above, we use in this work the same symbols for time-dependent quantities and their zeroth-order time-independent parts when unambiguous. It is to be understood that all relations containing perturbation parts are linear with respect to the latter and generally hold to first order only (as it should be per definition for a linear-response theory).

Substituting the first expansion in Eq. (29) into the orthonormality property of the orbitals, Eq. (20), we obtain (to first order) the ‘integration-by-parts’ relation:

\[ \langle \delta \phi_k(r, t) | \phi_q(r) \rangle = -\langle \phi_k(r) | \delta \phi_q(r, t) \rangle, \quad k, q = 1, \ldots, M. \] (30)

Similarly, making use of the differential condition Eq. (19) of MCTDHX one immediately arrives at the orthogonality of the perturbed parts of the orbitals with respect to the ground-state manifold of orbitals, namely:

\[ \langle \phi_k(r) | u_q(r) \rangle = 0, \quad \langle \phi_k(r) | v^*_q(r) \rangle = 0, \quad k, q = 1, \ldots, M. \] (31)

This relation is obtained to first order for \( \omega > 0 \) \[96\]. Any relation between the perturbed parts of the orbitals themselves is to second order, and cannot be deduced from the differential condition Eq. (19) within linear response.

Analogous relations can be obtained for the expansion coefficients. Substituting the third expansion (either of the two) in Eq. (29) into the orthonormality property of the coefficients, Eq. (22), we obtain (to first order) the ‘integration-by-parts’ relation:

\[ (\delta C)^\dagger C = -C^\dagger \delta C. \] (32)
Similarly, making use of the differential condition for the coefficients, Eq. (26), one immediately arrives at the orthogonality of the perturbed parts of the coefficients with respect to the ground-state (vector of) coefficients:

\[ C^\dagger C_u = 0, \quad C^\dagger C_v^* = 0. \] (33)

Again, this relation is obtained to first order for \( \omega > 0 \). \[97\]

C. 0-th order: MCHB and MCHF

The linear-response equations are derived by substituting the ansatz Eq. (29) into the MCTDHX equations of motion. To zeroth order one simply obtains the MCHX system itself which we repeat for reference:

\[ \hat{P} \sum_{q=1}^{M} [\rho_{kq} \hat{h} + \sum_{s,l=1}^{M} \rho_{kstl} \hat{W}_{sl}] |\phi_q\rangle = 0, \quad k = 1, \ldots, M, \]

\[ P_C H C = 0 \quad \iff \quad C H^{(-\epsilon)} = 0. \] (34)

When possible or instructive, we will avoid writing the Lagrange multipliers explicitly.

D. 1-st order: Orbitals

We now derive separately the first order (linear-response) equations emanating from the orbitals’ and from the coefficients’ (subsequent Subsec. [ITE]) parts of MCTDHX. This is done by formally taking the variation of the MCTDHX equations of motion. We find from Eq. (17) for the perturbed orbitals that:

\[ \delta \hat{P} \sum_{q=1}^{M} [\rho_{kq} \hat{h} + \sum_{s,l=1}^{M} \rho_{kstl} \hat{W}_{sl}] |\phi_q\rangle + \]

\[ + \hat{P} \sum_{q=1}^{M} [\delta \rho_{kq} \hat{h} + \rho_{kq} \delta \hat{h} + \sum_{s,l=1}^{M} (\delta \rho_{kstl} \hat{W}_{sl} + \rho_{kstl} \delta \hat{W}_{sl})] |\phi_q\rangle + \]

\[ + \hat{P} \sum_{q=1}^{M} [\rho_{kq} \hat{h} + \sum_{s,l=1}^{M} \rho_{kstl} \hat{W}_{sl}] |\delta \phi_q\rangle = \]

\[ = i \sum_{q=1}^{M} \rho_{kq} |\delta \phi_q\rangle, \quad k = 1, \ldots, M. \] (35)
Note that a term with $\delta \rho_{kq}$ in front of the time derivative is to second order and hence does not enter here.

The evaluation of the first term in Eq. (35) employs the derived ‘integration-by-parts’ relation Eq. (30). Thus we find:

$$\delta \hat{P} \sum_{q=1}^{M} [\rho_{kq} \hat{h} + \sum_{s,l=1}^{M} \rho_{kstq} \hat{W}_{st}] |\phi_q \rangle = -\hat{P} \sum_{q=1}^{M} \mu_{kq} |\delta \phi_q \rangle, \quad k = 1, \ldots, M. \tag{36}$$

Using Eq. (31) and similarly to Eq. (36), the projection operator $\hat{P}$ can also be reinstated in front of the time derivative on the right-hand side of the linear-response system Eq. (35). This will be used later on. With Eq. (36), the variation in Eq. (35) simplifies and we get:

$$\hat{P} \sum_{q=1}^{M} [\delta \rho_{kq} \hat{h} + \rho_{kq} \delta \hat{h} + \sum_{s,l=1}^{M} (\delta \rho_{kstq} \hat{W}_{st} + \rho_{kstq} \delta \hat{W}_{st})] |\phi_q \rangle +$$

$$+\hat{P} \sum_{q=1}^{M} [\rho_{kq} (\hat{h} - \omega) - \mu_{kq} + \sum_{s,l=1}^{M} \rho_{kstq} \hat{W}_{st}] |\delta \phi_q \rangle =$$

$$= i \sum_{q=1}^{M} \rho_{kq} |\delta \dot{\phi}_q \rangle, \quad k = 1, \ldots, M. \tag{37}$$

Eq. (37) is the generic form for the linear-response equations of the orbitals, formally for $\omega > 0$. The derivation for time-independent perturbations is beyond our scope here. In case of bosons and for a single orbital, that is for $M = 1$, Eq. (37) boils down to a particle-conserving version of the Bogoliubov–de Gennes equations [24]. When the number of fermions equals the number of orbitals, namely $M = N$, Eq. (37) boils down to the random-phase approximation, see in this context, e.g., Ref. [12]. The orbitals’ projector guarantees that this is an orthogonal version of the random-phase approximation, where all orbitals’ perturbations are automatically orthogonal to the ground-state determinant.

Let us now proceed and derive the explicit equations for the perturbed orbitals, namely substitute the ansatz Eq. (29) into the result Eq. (37). The first-order equation associated with $e^{-\im \omega t}$ is given by:

$$\hat{P} \sum_{q=1}^{M} [\delta \rho_{kq} |e^{-\im \omega t} \hat{h} + \sum_{s,l=1}^{M} (\delta \rho_{kstq} |e^{-\im \omega t} \hat{W}_{st} + \rho_{kstq} \delta \hat{W}_{st}|e^{-\im \omega t})] |\phi_q \rangle +$$

$$+\hat{P} \sum_{q=1}^{M} [\rho_{kq} (\hat{h} - \omega) - \mu_{kq} + \sum_{s,l=1}^{M} \rho_{kstq} \hat{W}_{st}] |\delta \phi_q \rangle =$$

$$= -\hat{P} \sum_{q=1}^{M} (\rho_{kq} \hat{\delta} + \sum_{s,l=1}^{M} \rho_{kstq} \{\hat{g} \} \delta_{st}) |\phi_q \rangle, \quad k = 1, \ldots, M, \tag{38}$$
where \( \{ \hat{g}^\dagger \}_s l = \int dr' \phi_s^*(r') \hat{g}^\dagger (r - r') \phi_l(r') \). Since in view of the above it is allowed, we for now put the \( \omega \)-term inside, i.e., under the projector.

We can now proceed and substitute the perturbed quantities under the variation in Eq. (38). This leads to the following ingredients for the \( e^{-i\omega t} \) equation:

\[
\hat{W}_{sl} \quad \Rightarrow \quad \delta \hat{W}_{sl}|_{e^{-i\omega t}} = \hat{W}_{l s l} + \hat{W}_{s l s},
\]
\[
\rho_{k q} = C^\dagger \cdot \hat{\rho}_{k q} \quad \Rightarrow \quad \delta \rho_{k q}|_{e^{-i\omega t}} = (C^\dagger \hat{\rho}_{k q})^t \cdot C_v + (C^\dagger \hat{\rho}_{k q})^\dagger \cdot C_u,
\]
\[
\rho_{k_{slq}} = C^\dagger \cdot \hat{\rho}_{k_{slq}} \quad \Rightarrow \quad \delta \rho_{k_{slq}}|_{e^{-i\omega t}} = (C^\dagger \hat{\rho}_{k_{slq}})^t \cdot C_v + (C^\dagger \hat{\rho}_{k_{lsq}})^\dagger \cdot C_u,
\]

(39)

where, to remind, the transformed coefficients \( \hat{\rho}_{k q} = \{ C_{\vec{n} k q} \} \) are defined as \( \hat{\rho}_{k q} \sum_{\vec{n}} C_{\vec{n} k q} | \vec{n}; t \rangle \equiv \sum_{\vec{n}} C_{\vec{n} k q} | \vec{n}; t \rangle \), and similarly for \( \hat{\rho}_{k_{slq}} = \{ C_{\vec{n} k_{slq}} \} \). The final result reads:

\[
\hat{P} \sum_{q=1}^M \left[ \left( \left( C^\dagger \hat{\rho}_{k q} \right)^t \cdot C_v + (C^\dagger \hat{\rho}_{k q})^\dagger \cdot C_u \right) \hat{h} + \sum_{s,l=1}^M \left( \left( C^\dagger \hat{\rho}_{k_{slq}} \right)^t \cdot C_v + (C^\dagger \hat{\rho}_{k_{lsq}})^\dagger \cdot C_u \right) \hat{W}_{sl} \right] | \phi_q \rangle +
\]
\[
+ \hat{P} \sum_{q=1}^M \left[ \rho_{k q} (\hat{h} - \omega) - \mu_{k q} + \sum_{s,l=1}^M \rho_{k_{slq}} (\hat{W}_{sl} \pm \hat{K}_{sl}) \right] | u_q \rangle + \hat{P} \sum_{q,s,l=1}^M \rho_{k_{qls}} \hat{K}_{rs} | v_q \rangle =
\]
\[
= - \hat{P} \sum_{q=1}^M (\hat{\rho}_{k q} \hat{f}^\dagger + \sum_{s,l=1}^M \rho_{k_{slq}} \{ \hat{g}^\dagger \}_s l ) | \phi_q \rangle, \quad k = 1, \ldots, M,
\]

(40)

where the \( \pm \) sign refers to bosons or fermions, respectively, and the exchange operator is introduced and defined as:

\[
\hat{K}_{sl} = \int dr' \phi_s^*(r') \hat{W}(r - r') \hat{P}_{rr'} \phi_l(r'), \quad \hat{K}_{sl} f(r) \equiv \hat{W}_{sf} \phi_l(r)
\]

(41)

with \( \hat{P}_{rr'} \) permuting the coordinates of two particles. We emphasize that MCTDHX and MCHX are formulated with local potentials only, see Eq. (18). For their linear-response theories, the exchange potentials, Eq. (41), with the perturbed orbitals appear, see the term \( \pm \hat{K}_{sl} | u_q \rangle \) in Eq. (40). Furthermore, the \( \pm \) in front of this exchange term implies that the dependence on the particles’ statistics appears explicitly in linear response, also see further below, unlike the implicit dependence (“invariance”) on particles’ statistics of MCTDHX and MCHX, see Ref. [45].
The first-order equation associated with $e^{i\omega t}$ is given by:

$$
\hat{P}^* \sum_{q=1}^{M} \left[ \{\delta \rho_{kq}\}_{e^{i\omega t}} \hat{h}^* + \sum_{s,l=1}^{M} \{\delta \rho_{klq}\}_{e^{i\omega t}} \hat{W}_{ls} + \rho_{qlsk} \{\delta \hat{W}_{st}\}_{e^{i\omega t}} \right] |\phi_q^*\rangle + 
+ \hat{P}^* \sum_{q=1}^{M} [\rho_{kq}(\hat{h}^* + \omega) - \mu_{kq} + \sum_{s,l=1}^{M} \rho_{qlsk} \hat{W}_{ls}] |v_q\rangle = 
= -\hat{P}^* \sum_{q=1}^{M} (\rho_{kq} \hat{f}^* + \sum_{s,l=1}^{M} \rho_{qlsk} \{\hat{g}^\dagger\}_{ls}) |\phi_q^*\rangle, \quad k = 1, \ldots, M, \quad (42)
$$

where the hermiticity of the Lagrange multipliers' matrix $\mu_{kq}^* = \mu_{kq}$ has been used. Again, we for now put the $\omega$-term under the projector in view of Eq. (31).

Similarly to the above treatment we obtain the following ingredients for the $e^{i\omega t}$ equation [utilizing the $e^{-i\omega t}$ ones, see Eq. (39)]:

\[ [\delta \hat{W}_{st}|_{e^{i\omega t}}]^\dagger \iff \delta \hat{W}_{ls}|_{e^{-i\omega t}}, \]
\[ [\delta \rho_{kq}|_{e^{i\omega t}}]^\dagger \iff \delta \rho_{qk}|_{e^{-i\omega t}}, \]
\[ [\delta \rho_{klq}|_{e^{i\omega t}}]^\dagger \iff \delta \rho_{qlsk}|_{e^{-i\omega t}}, \quad (43) \]

where, of course, $[\delta \hat{h}|_{e^{i\omega t}}]^\dagger = \delta \hat{h}|_{e^{-i\omega t}}$ holds \[98\]. The final result reads:

$$
\hat{P}^* \sum_{q=1}^{M} \left[ \{\delta \rho_{kq}\}_{e^{i\omega t}}^\dagger \cdot \hat{C}_v + \{\delta \rho_{kq}\}_{e^{i\omega t}}^\dagger \cdot \hat{C}_u \right] \hat{h}^* + \sum_{s,l=1}^{M} \left\{ \{\delta \rho_{qlsk}\}_{e^{i\omega t}}^\dagger \cdot \hat{C}_v + \{\delta \rho_{qlsk}\}_{e^{i\omega t}}^\dagger \cdot \hat{C}_u \right\} \hat{W}_{ls} |\phi_q^*\rangle + 
+ \hat{P}^* \sum_{q=1}^{M} [\rho_{kq}(\hat{h}^* + \omega) - \mu_{kq} + \sum_{s,l=1}^{M} \rho_{qlsk} \hat{W}_{ls}] |v_q\rangle + \hat{P}^* \sum_{q,s,l=1}^{M} \rho_{qlsk} \hat{K}_{st} |u_q\rangle = 
= -\hat{P}^* \sum_{q=1}^{M} (\rho_{kq} \hat{f}^* + \sum_{s,l=1}^{M} \rho_{qlsk} \{\hat{g}^\dagger\}_{ls}) |\phi_q^*\rangle, \quad k = 1, \ldots, M. \quad (44)
$$

Observe and compare Eqs. (44) and (40) for how they are related. We will return to this matter in Subsec. III F.

### E. 1-st order: Coefficients

Next, we now move to the perturbed coefficients. We find from Eq. (17) for the perturbed coefficients that:

$$
\delta \hat{C}^H + C^{\delta H} = \varepsilon \delta \hat{C} + i \delta \hat{C} \iff \delta \hat{C}^{\delta H - \varepsilon} + C^{\delta H} = i \delta \hat{C}. \quad (45)
$$
Furthermore, from the “fully projected” MCTDHX equations of motion, Eq. (24), we find for the perturbed coefficients that:

\[ \delta \mathcal{P}_C \mathcal{C}^\hat{H} + \mathcal{P}_C (\delta \mathcal{C}^\hat{H} + \mathcal{C}^\delta \hat{H}) = i \delta \mathcal{C} \quad \iff \quad \mathcal{P}_C (\delta \mathcal{C}^{\hat{H} - \varepsilon} + \mathcal{C}^{\delta \hat{H}}) = i \delta \mathcal{C}, \quad (46) \]

where use has been made in the relation [compare to Eq. (36)]:

\[ \delta \mathcal{P}_C \mathcal{H} \mathcal{C} = -\varepsilon \mathcal{P}_C \delta \mathcal{C} \quad \iff \quad \delta \mathcal{P}_C \mathcal{C}^\hat{H} = \mathcal{P}_C \delta \mathcal{C}^{\varepsilon}. \quad (47) \]

From now on we will use the “fully projected” MCTDHX theory in order to derive the coefficients’ equation of motion. The results from the standard MCTDHX equations of motion can generally be obtained from the following by dropping \( \mathcal{P}_C \) therein.

We will also be needing the Hamiltonian and its variation, expressed in terms of density operators \([99]\):

\[ \hat{\mathcal{H}} = \sum_{k,q=1}^{M} [h_{kq} \hat{\rho}_{kq} + \frac{1}{2} \sum_{s,l=1}^{M} W_{ksql} \hat{\rho}_{kslq}] \quad (48) \]

and

\[ \delta \hat{\mathcal{H}} = \sum_{k,q=1}^{M} [(h_{\delta kq} + h_{k\delta q} + \{ \delta h \}_{kq}) \hat{\rho}_{kq} + \]

\[ + \frac{1}{2} \sum_{s,l=1}^{M} (W_{\delta ksqt} + W_{k\delta sqt} + W_{ksdq} + W_{ksdq} + \{ \delta W \}_{ksql}) \hat{\rho}_{kslq}] = \]

\[ = \sum_{k,q=1}^{M} [(\{ h^* \}_{q^* \delta k} + h_{k\delta q} + \{ \delta h \}_{kq}) \hat{\rho}_{kq} + \sum_{s,l=1}^{M} (W_{sq^* \delta k} + W_{skl} + \frac{1}{2} \{ \delta W \}_{ksql}) \hat{\rho}_{kslq}] . \quad (49) \]

Note that for current convenience we write the perturbed part of the Hamiltonian (i.e., the perturbing fields) only in its variation part, also see below.

Let us now proceed and derive the explicit equations for the perturbed coefficients, namely substitute the ansatz Eq. (29) into the result Eq. (46). The first-order equation associated with \( e^{-i\omega t} \) is given by:

\[ \mathcal{P}_C \mathcal{C}^{\hat{\mathcal{H}} + \varepsilon - \hat{\mathcal{H}}} = \mathcal{P}_C \mathcal{C}^{\delta \hat{\mathcal{H}} |_{e^{-i\omega t}}}, \quad (50) \]

\[ \delta \hat{\mathcal{H}} |_{e^{-i\omega t}} = \sum_{k,q=1}^{M} [(\{ h^* \}_{q^* \nu k} + h_{k\nu q} + \{ f^\dagger \}_{kq}) \hat{\rho}_{kq} + \sum_{s,l=1}^{M} (W_{sq^* \nu k} + W_{skl} + \frac{1}{2} \{ g^\dagger \}_{ksql}) \hat{\rho}_{kslq}] . \]
The first-order equation associated with $e^{+i\omega t}$ is given by:

$$P^*_C C^\varepsilon -\omega - \hat{H}^* = P^*_C (C^*)^\dagger \{\delta \hat{H}_{|e^{+i\omega t}}\}^*, \quad (51)$$

$$\hat{H}^* = \sum_{k,q=1}^{M} \left[ h_{kq} \hat{\rho}_{kq} + \frac{1}{2} \sum_{s,l=1}^{M} W_{lqsk} \hat{\rho}_{kqlq} \right],$$

$$\{\delta \hat{H}_{|e^{+i\omega t}}\}^* = \sum_{k,q=1}^{M} \left[ (h_{kq} + \{h^*\}^q_{k^*}) \hat{\rho}_{kq} + \sum_{s,l=1}^{M} (W_{lqsk} + W_{l^*q^*s} + \frac{1}{2} \{g^\dagger\}_{lqsk}) \hat{\rho}_{kqlq} \right].$$

Here $\varepsilon$ is the MCHX ground-state energy and is, of course, real valued. Note that the complex conjugate $P^*_C$ appears for the perturbed coefficients as does the complex conjugate $\hat{P}^*$ for the perturbed orbitals, see Eq. (44). In Eq. (51) we introduced for convenience the star ($\star$) of a second-quantized operator, $\hat{O}^\star$, which is related to the original operator $\hat{O}$ as follows: (i) take the complex conjugate of the one-body and two-body matrix elements with respect to the orbitals, and (ii) do not take the hermitian conjugate of the density operators. The star operation comes naturally when utilizing complex conjugation within mapping in Fock space. Of course, since the Hamiltonian is hermitian, the usual hermiticity holds for the $e^{\mp i\omega t}$ variations of the Hamiltonian:

$$\{\delta \hat{H}_{|e^{+i\omega t}}\}^\dagger = \{\delta \hat{H}_{|e^{-i\omega t}}\}. \quad (52)$$

We remind the notation for the transformed coefficients in Eqs. (50) and (51) where, e.g., $C^\varepsilon -\omega - \hat{H}^* = \{C^\varepsilon -\omega - \hat{H}^*\}$ is defined as follows: $(\varepsilon - \omega - \hat{H}^*) \sum_\bar{n} C_{v,\bar{n}} |\bar{n}; t\rangle \equiv \sum_\bar{n} C^\varepsilon -\omega - \hat{H}^* |\bar{n}; t\rangle$ with $C_v = \{C_{v,\bar{n}}\}$.

**F. The linear-response matrix system and its formal solution**

We now proceed and cast the coupled linear-response system, Eqs. (40), (44), (50), and (51), into a matrix form. This is done in Subsec. III F 1 where we first post the main result and subsequently go through the individual ingredients that make it. With the linear-response matrix system we can formally solve the time-dependent many-body Schrödinger equation in linear response. This is done in Subsec. III F 2 where we first discuss the symmetry and other properties of the linear-response matrix system, and subsequently solve for the perturbed time-dependent orbitals and coefficients, and the MCTDHX wavefunction in linear response.
1. Casting the linear-response equations into a matrix form

Since the responses of the orbitals and coefficients are coupled, the central framework is to look at the system’s response space as a combined orbital–coefficient response space; also see in this respect Ref. [71]. Correspondingly, we define the combined vector of length $2(M + N_{\text{conf}})$:

$$\begin{pmatrix}
  u \\
  v \\
  C_u \\
  C_v
\end{pmatrix}, \quad u = \{|u_q\}_{q=1}^M, \ v = \{|v_q\}_{q=1}^M,$$

which collects all response amplitudes together for a given perturbed wavefunction.

Within this combined orbital–coefficient response space, the final result for the linear-response working matrix equation is:

$$(\mathbf{L} - \omega) \begin{pmatrix} u \\ v \\ C_u \\ C_v \end{pmatrix} = \mathbf{R}.$$  \hspace{1cm} (54)

The linear-response matrix $\mathbf{L}$ is explicitly assembled below, as does the vector $\mathbf{R}$ which collects the perturbing fields, see Eq. (29). We call Eq. (54) LR-MCTDHX theory, see in this context LR-MCTDHB [71].

To solve the LR-MCTDHX linear-response system Eq. (54), and hence the Schrödinger equation in linear response, we have to diagonalize the linear-response matrix $\mathbf{L}$ and find its excitations energies $\{\omega_k\}$ and eigenvectors:

$$\begin{pmatrix}
  u^k \\
  v^k \\
  C^k_u \\
  C^k_v
\end{pmatrix} = w_k \begin{pmatrix} u^k \\ v^k \\ C^k_u \\ C^k_v \end{pmatrix} \equiv w_k \mathbf{R}^k \hspace{1cm} (55)$$

This will be done in details in the following subsection [100].

We will now assemble and put together explicitly the associated response matrix. The
linear-response matrix is divided into four blocks:

\[
\mathcal{L} = \begin{pmatrix} \mathcal{L}_{oo} & \mathcal{L}_{oc} \\ \mathcal{L}_{co} & \mathcal{L}_{cc} \end{pmatrix}
\]

(56)

and, like the response vector Eq. (53), its dimension is \(2(M + N_{\text{conf}})\). The structure of \(\mathcal{L}\) represents the fact that the linear-response subspace is a combined space of orbitals and coefficients; the size of \(\mathcal{L}\) is twice as large as their combined sizes.

Each of the four blocks of \(\mathcal{L}\) is divided itself into four sub-matrices, representing the \(u\) and \(v\) quantities. Within each block, the four sub-matrices are linked between them as will be seen below. The orbital–orbital (oo) block reads:

\[
\mathcal{L}_{oo} = \begin{pmatrix} \mathcal{L}_{oo}^u & \mathcal{L}_{oo}^v \\ -(\mathcal{L}_{oo}^v)^* & -(\mathcal{L}_{oo}^u)^* \end{pmatrix}
\]

(57)

where \((k, q = 1, \ldots, M)\)

\[
\mathcal{L}_{oo}^u = \hat{\rho} \hat{h} - \mu + \Omega \pm \kappa^1,
\]

\[
\mathcal{L}_{oo}^v = \kappa^2 = \{\kappa^2_{kq}\} = \left\{ \sum_{s,l=1}^{M} \rho_{kqls} \hat{K}_{ls} \right\}.
\]

(58)

Inspecting of the block \(\mathcal{L}_{oo}\) reveals that the following further relations between its sub-matrices hold:

\[(\mathcal{L}_{oo}^u)^\dagger = (\mathcal{L}_{oo}^u), \quad (\mathcal{L}_{oo}^v)^t = (\mathcal{L}_{oo}^v).\]

(59)

The proof, especially with the non-local parts \(\kappa^1\) and \(\kappa^2\), follows straightforwardly by multiplying each sub-matrix with the identity (in the super-vector subspace of orbitals) from its left and right sides.

The orbital–coefficient (oc) block reads:

\[
\mathcal{L}_{oc} = \begin{pmatrix} \mathcal{L}_{oc}^u & \mathcal{L}_{oc}^v \\ -(\mathcal{L}_{oc}^v)^* & -(\mathcal{L}_{oc}^u)^* \end{pmatrix}
\]

(60)

where \((k = 1, \ldots, M)\)

\[
\mathcal{L}_{oc}^u = \left\{ \sum_{q=1}^{M} \hat{h}(C^\dagger \phi_q) + \sum_{s,l=1}^{M} \hat{W}_{sl}(C^\dagger \phi_q) \right\},
\]

\[
\mathcal{L}_{oc}^v = \left\{ \sum_{q=1}^{M} \hat{h}(C^\dagger \phi_q) + \sum_{s,l=1}^{M} \hat{W}_{sl}(C^\dagger \phi_q) \right\}.
\]

(61)
The coefficient–orbital (co) block reads:

\[
\mathcal{L}_{co} = \begin{pmatrix}
\mathcal{L}_{co}^u & \mathcal{L}_{co}^v \\
-(\mathcal{L}_{co}^v)^* & -(\mathcal{L}_{co}^u)^*
\end{pmatrix},
\tag{62}
\]

where \((k = 1, \ldots, M)\)

\[
\mathcal{L}_{co}^u = \left\{ \sum_{q=1}^{M} \langle \phi_q | \left( C\hat{\rho}_{qk} \right) \right. \hat{h} + \sum_{s,l=1}^{M} \left( C\hat{\rho}_{qsk} \right) W_{ls} \left. \right| \right\},
\]

\[
\mathcal{L}_{co}^v = \left\{ \sum_{q=1}^{M} \langle \phi_q^* | \left( C\hat{\rho}_{qk}^* \right) \right. \hat{h}^* + \sum_{s,l=1}^{M} \left( C\hat{\rho}_{qsk}^* \right) W_{sl} \left. \right| \right\}.
\tag{63}
\]

Inspection the sub-matrices of \(\mathcal{L}_{oc}\) and \(\mathcal{L}_{co}\) reveals the following relation between the two off-diagonal rectangular blocks of the response matrix \(\mathcal{L}\):

\[
(\mathcal{L}_{oc}^u)^\dagger = (\mathcal{L}_{co}^u), \quad (\mathcal{L}_{oc}^v)^t = (\mathcal{L}_{co}^v).
\tag{64}
\]

The proof is visualized directly by multiplying each sub-matrix with the identity (in the super-vector subspace of orbitals) and the identity in the subspace of coefficients, either from its left or right side, respectively. The relation in Eq. (64) can be used to reduce the computational effort in computing the response matrix, since the two off-diagonal blocks of \(\mathcal{L}\) do not need to be computed independently (see in this respect Ref. [71]). Finally, the coefficient–coefficient (cc) block reads:

\[
\mathcal{L}_{cc} = \begin{pmatrix}
(\cdot)^{\hat{H}-\varepsilon} & 0_c \\
0_c & (\cdot)^{\varepsilon-\hat{H}^*}
\end{pmatrix},
\tag{65}
\]

where \(0_c\) and \(1_c\) (see below) are zero and unit matrices of dimension \(N_{\text{conf}}\). The \((\cdot)\) symbol means that the operation on the response-coefficients’ part is performed within the mapping of coefficients [80], and the \(\varepsilon\) of the second-quantized Hamiltonian is defined in Eq. [51].

We will also be needing the orbitals–coefficients combined projector \(\mathcal{P}\) and combined metric \(\mathcal{M}\) matrices:

\[
\mathcal{P} = \begin{pmatrix}
\mathcal{P}_o & 0_{oo} \\
0_{oc} & \mathcal{P}_c
\end{pmatrix}, \quad \mathcal{P}_o = \begin{pmatrix}
\mathcal{P} & 0_o \\
0_o & \mathcal{P}^*
\end{pmatrix}, \quad \mathcal{P} = \hat{\mathcal{P}} 1_o, \quad \mathcal{P}_c = \begin{pmatrix}
\mathcal{P}_C & 0_c \\
0_c & \mathcal{P}_C^*
\end{pmatrix},
\tag{66}
\]

where \(1_o\) and \(0_o\) are unit and zero matrices of dimension \(M\), \(0_{oc}\) is a zero rectangular matrix of dimension \(M \times N_{\text{conf}}\), and \(0_{co} = (0_{oc})^t\). Similarly:

\[
\mathcal{M} = \begin{pmatrix}
\rho_o & 0_{oc} \\
0_{co} & 1_{2c}
\end{pmatrix}, \quad \rho_o = \begin{pmatrix}
\rho & 0_o \\
0_o & \rho^*
\end{pmatrix},
\tag{67}
\]
where $1_{2c}$ is a unit matrix of dimension $2N_{conf}$. Clearly, in view of the orthogonality relations Eqs. (31,33) every response vector satisfies:

\[
P \begin{pmatrix} u \\ v \\ C_u \\ C_v \end{pmatrix} = \begin{pmatrix} u \\ v \\ C_u \\ C_v \end{pmatrix},
\]

i.e., it lives in the complementary space of the ground-state wavefunction.

Finally, we collect the perturbing fields in the vector:

\[
M^{+\frac{1}{2}} R = M^{+\frac{1}{2}} \begin{pmatrix} -\hat{f}\\ -\hat{f}^* \end{pmatrix} + M^{-\frac{1}{2}} \begin{pmatrix} -\Omega g \phi \\ -\Omega g^* \phi^* \end{pmatrix},
\]

where $\phi = \{ |\phi_k\rangle \}, k = 1, \ldots, M$ is a column vector and $\Omega_g = \{ \Omega_{g,kq} \} = \{ \sum_{s,l=1}^{M} \rho_{kslq} \{ \hat{g}^\dagger \} \} \} a square matrix of dimension $M$.

With these ingredients at hand and inserting $P$ to the right (making use of the orthogonality of the response, Eq. (68), in the linear-response system or, equivalently, that initially $P$ does not multiply the $\omega$ term; see Ref. [71]), Eqs. (40), (44), (50), and (51) can be cast into the form:

\[
M^{+\frac{1}{2}} \left( PM^{-\frac{1}{2}} LM^{-\frac{1}{2}} P \right) - \omega \begin{pmatrix} u \\ C_u \\ v \\ C_v \end{pmatrix} \begin{pmatrix} u \\ C_u \\ v \\ C_v \end{pmatrix} = M^{+\frac{1}{2}} \left( PM^{+\frac{1}{2}} R \right).
\]

Multiply from the left by $M^{-\frac{1}{2}}$ and (to avoid cumbersome notation) using the assignments:

\[
\{ PM^{-\frac{1}{2}} LM^{-\frac{1}{2}} P \} \rightarrow L, \quad \{ PM^{+\frac{1}{2}} R \} \rightarrow R, \quad M^{+\frac{1}{2}} \begin{pmatrix} u \\ C_u \\ v \\ C_v \end{pmatrix} \rightarrow \begin{pmatrix} u \\ v \\ C_u \\ C_v \end{pmatrix},
\]

we arrive at the linear-response working matrix equation (54).
2. Solving the time-dependent identical-particle Schrödinger equation in linear response

We assume $L$ is diagonalizable and that all its eigenvalues are real (we have found this numerically to be the case for trapped repulsive bosons in their ground state \[71\]). Conversely and physically, when not all eigenvalues are real, an initial infinitesimal perturbation will grow up (within linear response) exponentially, implying that the system is unstable. We recall that, due to the projector matrix $P$ within $L$, the eigenvectors $\{R^k\}$ are in the complementary space, except for the zero excitations, assuming the case that, generally, the many-particle ground state is not degenerate.

Before we proceed and use the eigenvectors of $L$, Eq. (55), to solve the linear-response system Eq. (54), we briefly discuss the symmetries and other properties of the spectrum Eq. (55). The additional ingredient we need in order to analyze the solutions of the linear-response matrix $L$ are the ‘spin’ matrices:

$$\Sigma_1 = \begin{pmatrix} \Sigma_1^o & 0^c \\ 0^o & \Sigma_1^c \end{pmatrix} , \quad \Sigma_1^o = \begin{pmatrix} O_o & 1_o \\ 1_o & O_o \end{pmatrix} , \quad \Sigma_1^c = \begin{pmatrix} O_c & 1_c \\ 1_c & O_c \end{pmatrix} ,$$  

and

$$\Sigma_3 = \begin{pmatrix} \Sigma_3^o & 0^c \\ 0^o & \Sigma_3^c \end{pmatrix} , \quad \Sigma_3^o = \begin{pmatrix} 1_o & 0_o \\ 0_o & -1_o \end{pmatrix} , \quad \Sigma_3^c = \begin{pmatrix} 1_c & 0_c \\ 0_c & -1_c \end{pmatrix} .$$  

Making use of the $\Sigma_1$ matrix, Eq. (72), and examining each of the blocks of $L$, we find the symmetry property:

$$\Sigma_1 L \Sigma_1 = -(L)^* .$$  

Similarly, with the help of the $\Sigma_3$ matrix, Eq. (73), we find \[101\]:

$$\Sigma_3 L \Sigma_3 = (L)^\dagger .$$  

The symmetries Eqs. (74) and (75) lead to the following properties of the spectrum $\{\omega^k\}$ and eigenvectors $\{R^k\}$: From Eq. (74) we learn that $R^{-k} \equiv \Sigma_1 (R^k)^*$ is an eigenvector of $L$ with the eigenvalue $-(\omega_k)^*$, and from Eq. (75) we can construct the adjoint (or, left) eigenvectors $(L^k)^\dagger L = \omega^k (L^k)^\dagger$, where $L^k = \text{sng}^k \Sigma_3 R^k$ and $\text{sng}^k$ stands for the sign of the ‘scalar product’ $\{(R^k)^\dagger \Sigma_3 R^k\}$. The quantity $\text{sng}^k$ is formally introduced because the ‘scalar product’ is with the metric $\Sigma_3$ and thus can have a negative value, when the contribution from the $v$ terms is larger than the contribution from the $u$ ones. These allow us to obtain

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the orthogonality relations for eigenvectors with excitation index \( k \) and \( k' \):
\[
(L^k)^\dagger R^{k'} = \text{sgn}^k \left[ (u^k)^\dagger u^{k'} - (v^k)^\dagger v^{k'} + (C_u^k)^\dagger C_u^{k'} - (C_v^k)^\dagger C_v^{k'} \right] = \delta_{kk'},
\]
\[
(L^k)^\dagger R^{-k'} = \text{sgn}^k \left[ (u^k)^\dagger (v^{k'})^* - (v^k)^\dagger (u^{k'})^* + (C_u^k)^\dagger (C_v^{k'})^* - (C_v^k)^\dagger (C_u^{k'})^* \right] = 0.
\]

Finally, we can use these relations to write the resolution of the identity 1 and the spectral resolution of \( L \) from the positive (non-negative) sector of \( \{ R^k \} \). Thus we have:
\[
1 = \sum_{k=0}^{M^2} \{ R_0^k (L_0^k)^\dagger + R_0^{-k} (L_0^{-k})^\dagger \} + \sum_{k>M^2} \{ R^k (L^k)^\dagger + R^{-k} (L^{-k})^\dagger \},
\]
where \( L^{-k} = -\text{sgn}^k \Sigma_3 R^{-k} = -\text{sgn}^k \Sigma_3 \Sigma_1 (R^k)^* = \Sigma_1 (L^k)^* \). Note in comparison with \( L^k \) the minus sign in \( L^{-k} \), which emerges from the fact that \( \Sigma_3 \) and \( \Sigma_1 \) anti-commute. In Eq. (77) the first group of vectors are the zero-mode excitations. There are \( 2(M^2 + 1) \) such eigenvectors. \( M^2 \) eigenvectors are obtained by putting any of the \( M \) ground-state orbitals \( |\phi_q\rangle, q = 1, \ldots, M \) in any of the \( M \) entries of the \( u^k \) vector and 1 eigenvector is obtained by taking the ground-state vector of coefficients \( C \) as the vector \( C_u \). The other entries of \( R_0^k \) are all zero. This amount doubles on the account of \( \Sigma_1 \) and the negative (non-positive) sector. The second group of vectors in Eq. (77) are the non-zero-mode excitations. The respective summation index, \( k > M^2 \), indicates that we enumerate them after the group of zero-mode excitations for which the index of enumeration satisfies \( k \in [0, M^2] \). Finally, for \( L \) we have:
\[
L = \sum_{k>M^2} \omega_k \left\{ R^k (L^k)^\dagger - R^{-k} (L^{-k})^\dagger \right\}.
\]
It is instructive to compare the structure of Eqs. (77) and (78) to their analogs derived from the full Schrödinger equation, Eqs. (15) and (16).

We now return to Eq. (54) and employ the eigenvectors of \( L \) to solve it. To this end we expand the response amplitudes and perturbation as follows:
\[
\begin{pmatrix}
  u \\
  v \\
  C_u \\
  C_v
\end{pmatrix} = \sum_k c_k R^k = \sum_{k>M^2} \left[ c_k R^k + c_{-k} R^{-k} \right], \quad \mathcal{R} = - \sum_k \gamma_k R^k = - \sum_{k>M^2} \left[ \gamma_k R^k + \gamma_{-k} R^{-k} \right],
\]
\[
(79)
\]
where, to remind, $R^{-k} \equiv \Sigma_1(R^k)^*$. The zero-mode eigenvectors, as discussed above, do not contribute. Substituting Eq. (79) into Eq. (54) we find

$$
\begin{pmatrix}
u \\
v \\
w \end{pmatrix} = \sum_{k>M^2} \left[ \frac{\gamma_k}{\omega - \omega_k} R^k + \frac{\gamma_k}{\omega + \omega_k} R^{-k} \right],
$$

(80)

where the response weights are given explicitly by [see Eqs. (69) and (71)], $k > M^2$:

$$
\gamma_k = -(L^{k})^* R = \text{sgn}^k \left\{ (u^k)^* \left[ \rho^{\frac{1}{2}} f^\dagger + \rho^{-\frac{1}{2}} \Omega_g \right] \phi + (v^k)^* \left[ \rho^{\frac{1}{2}} f^* + \rho^{-\frac{1}{2}} \Omega_g^* \right] \phi^* \right. +
$$

$$
\left. + (C^k_u)^* \cdot C \{ \sum_{k,q=1}^{M} f^k q k \hat{p}_k + \frac{1}{2} \sum_{k,s,q,l=1}^{M} \{ g^q k k \hat{p}_k \} \} \right. +
$$

$$
\left. + (C^k_v)^* \cdot (C^*) \{ \sum_{k,q=1}^{M} f^k q k \hat{p}_k + \frac{1}{2} \sum_{k,s,q,l=1}^{M} \{ g^q k k \hat{p}_k \} \} \right\},
$$

(81)

$$
\gamma_{-k} = -(L^{-k})^* R = -\text{sgn}^k \left\{ (v^k)^t \left[ \rho^{\frac{1}{2}} f^\dagger + \rho^{-\frac{1}{2}} \Omega_g \right] \phi + (u^k)^t \left[ \rho^{\frac{1}{2}} f^* + \rho^{-\frac{1}{2}} \Omega_g^* \right] \phi^* \right. +
$$

$$
\left. + (C^k_u)^t \cdot C \{ \sum_{k,q=1}^{M} f^k q k \hat{p}_k + \frac{1}{2} \sum_{k,s,q,l=1}^{M} \{ g^q k k \hat{p}_k \} \} \right. +
$$

$$
\left. + (C^k_v)^t \cdot (C^*) \{ \sum_{k,q=1}^{M} f^k q k \hat{p}_k + \frac{1}{2} \sum_{k,s,q,l=1}^{M} \{ g^q k k \hat{p}_k \} \} \right\}.
$$

(81)

From the right-hand sides of Eq. (81) we see that the response weights combine the contributions from the responses of all orbitals and of all expansion coefficients. Note that $P$, see Eq. (71), falls out of the expression for the response weights.

Reinserting the expansion for the response amplitudes, Eq. (80), into the ansatz for the orbitals and expansion coefficients, Eq. (29), gives their time dependence in linear response:

$$
\phi(r, t) \approx \phi(r) + \delta \phi(r, t),
$$

$$
\delta \phi(r, t) = \sum_{k>M^2} \left\{ \left[ \gamma_k \{ \rho^k \} e^{-i\omega t} + \gamma_k^* \{ \rho^k \} e^{i\omega t} \right] / (\omega - \omega_k) +
$$

$$
+ \left[ \gamma_{-k} \{ \rho^k \} e^{i\omega t} + \gamma_{-k}^* \{ \rho^k \} e^{-i\omega t} \right] / (\omega + \omega_k) \right\},
$$

$$
C(t) \approx C + \delta C(t),
$$

$$
\delta C(t) = \sum_{k>M^2} \left\{ \left[ \gamma_k C_u^k e^{-i\omega t} + \gamma_k^* C_u^k e^{i\omega t} \right] / (\omega - \omega_k) +
$$

$$
+ \left[ \gamma_{-k} C_v^k e^{i\omega t} + \gamma_{-k}^* C_v^k e^{-i\omega t} \right] / (\omega + \omega_k) \right\},
$$

(82)
with \( \delta \phi(r, t) = \{ \delta \phi_j(r, t) \}, j = 1, \ldots, M \). Thus, the orbitals and the expansion coefficients show the largest response at the frequencies \( \{ \pm \omega_k \} \). Moreover, the response at a given frequency \( \omega_k \) is not necessarily equally strong for all the orbitals \( \phi(r, t) \) and similarly for all the expansion coefficients \( C(t) \). The reason is because the components of the response amplitudes \( u^k, v^k, C^k_u, \) and \( C^k_v \) are not individually normalized, but rather the whole amplitude vector is, see Eq. (76).

Finally, from Eq. (82) the time-dependent many-particle wavefunction is given in linear response by:

\[
\Psi(t) \approx \sum_{\vec{n}} C_{\vec{n}} |\vec{n}\rangle + \sum_{\vec{n}} \delta C_{\vec{n}}(t) |\vec{n}\rangle + \sum_{\vec{n}} C_{\vec{n}} \left[ \sum_{j=1}^{M} (\pm 1)^{\sum_{l=j+1}^{M} n_l} \sqrt{n_j} \sqrt{\langle \delta \phi_j(r, t) | \delta \phi_j(r, t) \rangle} |n_1, \ldots, n_j - 1, \ldots, n_M, 1_{M+1}; t \rangle \right],
\]

where \( |1_{M+1}; t \rangle \) is associated with the (unnormalized) time-dependent response orbital \( \delta \phi_j(r, t) \). The response orbitals \( \{ \delta \phi_j(r, t) \} \) need not be orthogonal to each other, unlike their orthogonality with the ground-state orbitals which originates from the orbital differential condition Eq. (19). This concludes our derivation of LR-MCTDHB and LR-MCTDHF, in a unified manner and representation.

### IV. LINEAR RESPONSE IN THE MULTICONFIGURATIONAL TIME-DEPENDENT HARTREE FRAMEWORK FOR DISTINGUISHABLE DEGREES-OF-FREEDOM

This section deals with systems of distinguishable degrees-of-freedom and – starting from the MCTDH propagation theory – it develops the corresponding linear-response theory, which we denote by LR-MCTDH, based on the knowhow of the previous section, Sec. III, and some new ingredients.

#### A. Quick derivation of MCTDH: Basic and new ingredients and notations

Let us have \( j = 1, \ldots, Q \) in-general distinguishable degrees-of-freedom which we label by the generalized coordinates \( r_1, \ldots, r_Q \). Each degree-of-freedom is expanded by \( n_j = 1, \ldots, M_j \) orthonormal time-dependent orbitals. Within a concise (yet clear) notation, the
many-particle wavefunction takes on the following appearance:

$$|\Psi(t)\rangle = \sum_{\vec{n}} C_{\vec{n}}|\vec{n}; t\rangle = \sum_{\vec{n}[j]} \sum_{n_j=1}^{M_j} C_{\vec{n}[j]n_j}|\vec{n}[j]; t\rangle|n_j; t\rangle, \quad \forall j. \quad (84)$$

The left-hand side of Eq. (84) looks the same as for identical particles, but the meaning is, of course, different. Namely, the $|\vec{n}; t\rangle$ are configurations–Hartree products, $|\vec{n}; t\rangle = \prod_{j=1}^{Q} |n_j; t\rangle$, not permanents or determinants.

The generic Hamiltonian of the $Q$ coupled degrees-of-freedom is:

$$\hat{H} = \sum_{j=1}^{Q} \hat{h}_j(r_j) + \hat{W}(r_1, \ldots, r_Q). \quad (85)$$

The coupling–interaction part is written generically. For instance, it can be comprised of few-body ingredients or, in the general case, couple all $Q$ degrees-of-freedom.

The action–functional reads (time-dependence of quantities is suppressed when unambiguous):

$$S = \int dt \left\{ \langle \Psi|\hat{H} - i\frac{\partial}{\partial t}|\Psi\rangle - \sum_{j=1}^{Q} \sum_{n_j=m_j}^{M_j} \rho_{n_jm_j}^j (\langle n_j|m_j\rangle - \delta_{n_jm_j}) - \varepsilon C\dagger C \right\}. \quad (86)$$

The MCTDH equations of motion for the time-dependent orbitals and expansion coefficients [28, 29] are readily derived by equating the variation of $S$ with respect to these quantities to zero. Thus one finds:

$$\dot{\rho}_{n_jm_j}^j = \sum_{n_j,m_j} \rho_{n_jm_j}^{j*} \hat{H}^j + \hat{\Omega}_{n_jm_j}^j |m_j\rangle \langle n_j|, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q,$$

$$\hat{H}(t)C(t) = i\frac{\partial C(t)}{\partial t}, \quad H_{\vec{n}\vec{n}'}(t) = \langle \vec{n}; t|\hat{H}|\vec{n}'; t\rangle, \quad (87)$$

where for each of the $j = 1, \ldots, Q$ degrees-of-freedom we define the reduced one-body density matrix:

$$\rho_{n_jm_j}^j = \sum_{\vec{n}[j]} C_{\vec{n}[j]n_j}^* C_{\vec{n}[j]m_j}, \quad n_j, m_j = 1, \ldots, M_j, \quad (88)$$

the mean-field operators:

$$\hat{\Omega}_{n_jm_j}^j = \sum_{\vec{n}[j], \vec{m}[j]} \rho_{\vec{n}\vec{m}} \hat{W}_{\vec{n}[j]\vec{m}[j]}, \quad n_j, m_j = 1, \ldots, M_j, \quad (89)$$
with \( \hat{W}_{\vec{n}[j]\vec{m}[j]} = \langle \vec{n}[j]|\hat{W}|\vec{m}[j] \rangle \), and \( \rho_{\vec{n}\vec{m}} = C_{\vec{n}}^*C_{\vec{m}} = \rho_{\vec{n}[j]\vec{m}[j]m_j} = C_{\vec{n}[j]|n_j}^*C_{\vec{m}[j]m_j}, \forall j \) is the (reduced) all-body density matrix. Finally, the projectors are defined as:

\[
P_j = 1 - \sum_{n'_j=1}^{M_j} |n'_j \rangle \langle n'_j |,
\]

We now proceed in the same manner as done in Sec. [III] above with the coefficients. The “fully projected” representation of MCTDH is obtained by assigning the coefficients as follows [see Eq. (23)]: \( C \to C e^{-i \int dt' C(t') H(t') C(t')} \). This results in:

\[
\hat{P}_j \sum_{m_j=1}^{M_j} [\rho^j_{n_j,m_j} \hat{h}^j + \hat{\Omega}^j_{n_j,m_j}] |m_j \rangle = i \sum_{m_j=1}^{M_j} \rho^j_{n_j,m_j} |\dot{m}_j \rangle, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q,
\]

\[
P(t)H(t)C(t) = i \frac{\partial C(t)}{\partial t},
\]

with \( P_C = 1 - CC^\dagger \). In Eq. (91) both the orbitals [28, 29] and the expansion coefficients satisfy the differential conditions:

\[
i \langle n_j | \dot{m}_j \rangle = 0, \quad n_j, m_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q, \quad iC^\dagger \dot{C} = 0,
\]

namely the evolution of the system’s wavefunction, Eq. (84), is completely orthogonal:

\[
i \langle \Psi(t) | \dot{\Psi}(t) \rangle = 0.
\]

Finally, from either Eq. (87) or Eq. (91) the time-independent MCH theory is obtained:

\[
\hat{P}_j \sum_{m_j=1}^{M_j} [\rho^j_{n_j,m_j} \hat{h}^j + \hat{\Omega}^j_{n_j,m_j}] |m_j \rangle = 0, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q, \iff
\]

\[
\sum_{m_j=1}^{M_j} [\rho^j_{n_j,m_j} \hat{h}^j - \mu^j_{n_j,m_j} + \hat{\Omega}^j_{n_j,m_j}] |m_j \rangle = 0, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q,
\]

\[
\mu^j_{n_j,m_j} = \langle m_j | \sum_{n'_j=1}^{M_j} [\rho^j_{n'_j,n_j} \hat{h}^j + \hat{\Omega}^j_{n'_j,n_j}] |n'_j \rangle = 0, \quad n_j, m_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q,
\]

\[
P_C HC = 0 \iff HC = \varepsilon C, \quad H_{\vec{n}\vec{n'}} = \langle \vec{n}|\hat{H}|\vec{n'} \rangle.
\]

Based on the linear response for indistinguishable particles of Sec. [III] and the above notation for MCTDH, we now proceed to derive LR-MCTDH. Only the essential formulas will be presented in detail.
B. Perturbation and variation

We derive the linear response of MCTDH using a small perturbation around the MCH solution, typically the ground state. We will consider both a one-body time-dependent perturbation, say an external field, and a generic “all-body” time-dependent perturbation, e.g., a change to the potential-energy hyper-surface between all degrees-of-freedom. The respective ansatz is as follows:

\[ |n_j; t⟩ \approx |n_j⟩ + |\delta n_j; t⟩, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q, \]

\[ |\delta n_j; t⟩ = |u_{n_j}⟩ e^{-i\omega t} + |v_{n_j}^*⟩ e^{+i\omega t}, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q, \]

\[ C(t) \approx [C + \delta C(t)] \quad \iff \quad C(t) \approx e^{-i\omega t}[C + \delta C(t)] \quad \text{(without coefficients’ projector)}, \]

\[ \delta C(t) = C_u e^{-i\omega t} + C_v e^{+i\omega t}, \]

\[ \delta \hat{h}^j(r_j, t) = \hat{f}^j_\dagger(r_j)e^{-i\omega t} + \hat{f}^j(r_j)e^{+i\omega t}, \quad j = 1, \ldots, Q, \]

\[ \delta \hat{W}(r_1, \ldots, r_Q, t) = \hat{g}^\dagger(r_1, \ldots, r_Q)e^{-i\omega t} + \hat{g}(r_1, \ldots, r_Q)e^{+i\omega t}. \] (94)

Here, \( \{|\delta n_j; t⟩\}; \quad j = 1, \ldots, Q \) and \( \delta C(t) \) are the perturbed parts of the orbitals and coefficients, respectively. The perturbed parts of the system’s wavefunction are all comprised of \( u \) and \( v \) contributions. The operators \( \{\hat{f}^j_\dagger(r_j)\}; \quad j = 1, \ldots, Q \) and \( \hat{g}(r_1, \ldots, r_Q) \) generate independently the one-body and all-body perturbations. It is instructive to contrast the perturbation ansatz Eq. (94) for distinguishable particles and the respective one for identical particles, Eq. (29). In particular, each of the \( j = 1, \ldots, Q \) distinct degrees-of-freedom can, in principle, be perturbed by a different one-body operator, \( \hat{f}_j \). We emphasize that despite the distinguishability, the whole coupled system responds as a whole, even if only, say the \( j_0 \)-th degree-of-freedom is perturbed, and the remaining ones are not, namely \( \hat{f}_{j \neq j_0} = 0 \) and \( \hat{g} = 0 \). As was used above, the perturbing frequency \( \omega \) is assumed to be non-zero. The Hamiltonian including the perturbation is hermitian.

In a similar manner like in Sec. III one can derive the orthogonality conditions between the perturbations and ground-state quantities (orbitals and coefficients). We will not repeat this step here.
C. The linear-response system and its formal solution

Our goal now is to develop the linear-response theory and solve the respective many-body Schrödinger equation in linear response. To assist the reader, we have divided the task to three. First, in Subsec. IV C 1 we linearize the MCTDH theory and derive separately the orbitals’ and coefficients’ linear-response equations. Then, in Subsec. IV C 2, we cast them into a matrix form, adapting the same strategy as done for identical particles above. Finally, in Subsec. IV C 3 after discussing the symmetries and other properties of the linear-response matrix system we solve for the perturbed time-dependent orbitals and coefficients, and the MCTDH wavefunction in linear response.

1. Linear-response equations

Substituting the ansatz Eq. (94) into the MCTDH equations of motion, either into the standard form, Eq. (87), or into the “fully projected” form, Eq. (91), we find to 0-th order the MCH equations themselves, see Eq. (93). For the linear-response (1-st order) equations we will proceed, as above, separately for the orbitals and for the coefficients. To derive the equations for the coefficients, we will work with the “fully projected” form, Eq. (91), namely employ explicitly the coefficients’ projector \( P_C \).

We find from either the standard, Eq. (87), or the “fully projected” form, Eq. (91), of MCTDH the following relation for the perturbed orbitals:

\[
\hat{P}_j \sum_{m_j=1}^{M_j} \left[ \delta \rho_{n_j m_j}^{j} \hat{h}^{j} + \rho_{n_j m_j}^{j} \delta \hat{h}^{j} + \delta \hat{\Omega}_{n_j m_j}^{j} \right] |m_j\rangle + \\
+ \hat{P}_j \sum_{m_j=1}^{M_j} \left[ \rho_{n_j m_j}^{j} \hat{h}^{j} - \rho_{n_j m_j}^{j} \hat{\mu}_{n_j m_j}^{j} + \hat{\Omega}_{n_j m_j}^{j} \right] |\delta m_j\rangle = \\
i \sum_{m_j=1}^{M_j} \rho_{n_j m_j}^{j} |\delta \hat{m}_j\rangle, \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q,
\] (95)

where we have used \( \delta \hat{P}_j \sum_{m_j=1}^{M_j} [\rho_{n_j m_j}^{j} \hat{h}^{j} + \hat{\Omega}_{n_j m_j}^{j} |m_j\rangle = -\hat{P}_j \sum_{m_j=1}^{M_j} \rho_{n_j m_j}^{j} |\delta m_j\rangle, n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q \). Eq. (95) is the generic form for the linear-response equations for the orbitals’ part of \( Q \) coupled degrees-of-freedom. In the same manner that for \( M_1 = \ldots = M_Q = 1 \) MCTDH boils down to the text-book time-dependent Hartree (TDH)
equations, see, e.g., Ref. [38], Eq. (95) boils down to the linear response of TDH (LR-TDH), which can readily be prescribed; we do not write explicitly the resulting equations here.

Let us proceed to the main steps in the derivation of the LR-MCTDH equations. We need to consider the variation leading to the $e^{+i\omega t}$ terms separately. This leads to the following ingredients for the $e^{-i\omega t}$ equation:

\[
\delta \tilde{W}_{\tilde{n}[j]\tilde{m}[j]}\bigg|_{e^{-i\omega t}} = \sum_{k \neq j}^{Q} (\tilde{W}_{\tilde{n}[j,k]\tilde{m}[j]v_{n_k}} + \tilde{W}_{\tilde{n}[j]\tilde{m}[j,k]u_{m_k}}),
\]

\[
\delta \tilde{\Omega}_{n,j,m} \bigg|_{e^{-i\omega t}} = \sum_{n[j],\tilde{m}[j]} (\{C^t\rho_{\tilde{n}\tilde{m}} \cdot C_v + C^t\rho_{\tilde{n}\tilde{m}} \cdot C_u\} \tilde{W}_{\tilde{n}[j]\tilde{m}[j]} + \rho_{\tilde{n}\tilde{m}} \delta \tilde{W}_{\tilde{n}[j]\tilde{m}[j]}\bigg|_{e^{-i\omega t}}),
\]

\[
\delta \rho_{n,j,m} \bigg|_{e^{-i\omega t}} = C^t\rho_{m_j,n_j} \cdot C_v + C^t\rho_{n_j,m_j} \cdot C_u,
\]

where we have used a tensor-product representation to work with the vector of coefficients and its response–variation explicitly, see Appendix [3]. In the above equation for the variation of $\tilde{W}_{\tilde{n}[j]\tilde{m}[j]}$ with the summation over $k \neq j$ it is implicitly taken that $n_k \in \tilde{n}[j], m_k \in \tilde{m}[j]$. With the ingredients in Eq. (96) we get the explicit first-order equation associated with the $e^{-i\omega t}$ term:

\[
\dot{P}_j \sum_{m_j=1}^{M_j} [\{C^t\rho_{m_j,n_j} \cdot C_v + C^t\rho_{n_j,m_j} \cdot C_u\} \dot{J}_j + \sum_{n[j],\tilde{m}[j]} (\{C^t\rho_{\tilde{n}\tilde{m}} \cdot C_v + C^t\rho_{\tilde{n}\tilde{m}} \cdot C_u\} \tilde{W}_{\tilde{n}[j]\tilde{m}[j]}|m_j) + \dot{P}_j \sum_{m_j=1}^{M_j} \rho_{n_j,m_j}(\dot{J}_j - \omega - \mu_{n_j,m_j} + \dot{J}_j)|u_{m_j}) + \dot{P}_j \sum_{k \neq j}^{Q} \sum_{m_k=1}^{M_k} \sum_{n[j],\tilde{m}[k]} \rho_{\tilde{n}\tilde{m}}(\dot{K}_{\tilde{n}[j]\tilde{m}[k]}|u_{m_k}) + \dot{K}_{\tilde{n}[j,k]\tilde{m}[k]}|v_{n_k}) = \]

\[
= -\dot{P}_j \sum_{m_j=1}^{M_j} (\rho_{n_j,m_j} \dot{J}_j + \sum_{n[j],\tilde{m}[j]} \rho_{\tilde{n}\tilde{m}}(\dot{g}^\dagger_{\tilde{n}[j]\tilde{m}[j]})|m_j), \quad n_j = 1, \ldots, M_j, \quad j = 1, \ldots, Q.
\]

Note the similarity and differences between Eq. (97) for distinguishable degrees-of-freedom and Eq. (10) for identical particles. In particular, the responses of different degrees-of-
freedom are coupled by the exchange-like potentials:

\begin{equation}
\hat{K}_{m[j]m[k]}|u_{m_k}\rangle \equiv \hat{W}_{m[j]m[j,k]u_m}|m_j\rangle, \quad \hat{K}_{m[j]m[k]}|v_{m_k}\rangle \equiv \hat{W}_{m[j,k]m[j]v_m}|m_j\rangle. \tag{98}
\end{equation}

We will return to this point below.

Similarly, the final result for the first-order equation associated with the \(e^{+i\omega t}\) term is given by:

\begin{align*}
\hat{P}_j^{*} \sum_{m_j=1}^{M_j} \left[ \{ C^t \rho_{n_j m_j} \cdot C_v + C^t \rho_{m_j n_j} \cdot C_u \} \{ \hat{h}^j \}^* + \\
+ \sum_{\tilde{m}[j], \tilde{m}[j]} \sum_{\tilde{m}[j], \tilde{m}[j]} \left( C^t \rho_{\tilde{m}\tilde{m}} \cdot C_v + C^t \rho_{\tilde{m}\tilde{m}} \cdot C_u \right) \hat{W}_{\tilde{m}[j] \tilde{m}[j]} |m_j^*\rangle + \\
+ \hat{P}_j^{*} \sum_{m_j=1}^{M_j} \left[ \rho_{\tilde{m}\tilde{m}} \left( \{ \hat{h}^j \}^* + \omega \right) - \mu_{m_j n_j} + \hat{\Omega}_j^j \right]|v_{m_j}\rangle + \\
= -\hat{P}_j^{*} \sum_{m_j=1}^{M_j} \left( \rho_{\tilde{m}\tilde{m}} \hat{f}_j^* + \sum_{\tilde{m}[j], \tilde{m}[j]} \rho_{\tilde{m}\tilde{m}} \left( \hat{g}_j^j \right)_{\tilde{m}[j] \tilde{m}[j]} |m_j^*\rangle \right), \quad n_j = 1, \ldots, M_J, \quad j = 1, \ldots, Q,
\end{align*}

where

\begin{equation}
\hat{K}_{\tilde{m}\tilde{m}[j]k}|u_{m_k}\rangle \equiv \hat{W}_{\tilde{m}[j] \tilde{m}[j,k]u_m}|m_j^*\rangle, \quad \hat{K}_{\tilde{m}[k]\tilde{m}[j]}|v_{m_k}\rangle \equiv \hat{W}_{\tilde{m}[j,k] \tilde{m}[j]v_m}|m_j^*\rangle. \tag{100}
\end{equation}

The connection between the \(e^{-i\omega t}\) equations stems from (the complex conjugation and) the relations:

\begin{align*}
[\delta \rho_{n_j m_j}^j |e^{+i\omega t}\rangle]^{\dagger} & \iff \delta \rho_{m_j n_j}^j |e^{-i\omega t}\rangle, \\
[\delta \rho_{\tilde{m}\tilde{m}} |e^{+i\omega t}\rangle]^{\dagger} & \iff \delta \rho_{\tilde{m}\tilde{m}} |e^{-i\omega t}\rangle, \\
[\delta \hat{W}_{\tilde{m}[j] \tilde{m}[j]}^j |e^{+i\omega t}\rangle]^{\dagger} & \iff \delta \hat{W}_{\tilde{m}[j] \tilde{m}[j]} |e^{-i\omega t}\rangle, \\
[\delta \hat{\Omega}_j^j |e^{+i\omega t}\rangle]^{\dagger} & \iff \delta \hat{\Omega}_j^j |e^{-i\omega t}\rangle.
\end{align*}

as for the identical-particle case, see Eq. (43).

How to boil down the LR-MCTDH equations when the all-body potential (and perturbations) are written as sums of products of one-body operators, a useful representation within the MCTDH algorithm [39, 40], is straightforward and will not be pursued here.
We now move to the perturbed coefficients, namely to the linear-response equations in 1-st order of the coefficients’ part. For the “fully projected” case (we will only treat it within LR-MCTDH) we find that:

\[
P_C[(\mathbf{H} - \varepsilon)\delta \mathbf{C} + (\delta \mathbf{H})\mathbf{C}] = i\delta \mathbf{C},
\]

just like Eq. (46) which utilized Eq. (47) in the indistinguishable-particle system. Using a tensor-product representation (see Appendix B) and in analogy to Eqs. (48) and (49) for identical particles, the Hamiltonian matrix:

\[
\mathbf{H} = \sum_{j=1}^{Q} \sum_{n_j, m_j=1}^{M_j} h_{n_j m_j}^j \mathbf{P}_{n_j m_j}^j + \sum_{\tilde{n}, \tilde{m}} W_{\tilde{n} \tilde{m}} \mathbf{P}_{\tilde{n} \tilde{m}},
\]

and its variation:

\[
\delta \mathbf{H} = \sum_{j=1}^{Q} \sum_{n_j, m_j=1}^{M_j} (h_{\tilde{n} \tilde{m}}^j + h_{n_j \delta m_j}^j + \{\delta h^j\}_{n_j m_j}) \mathbf{P}_{n_j m_j}^j + \sum_{\tilde{n}, \tilde{m}} (W_{\tilde{n} \tilde{m}} + W_{\tilde{n} \delta \tilde{m}} + \{\delta W\}_{\tilde{n} \tilde{m}}) \mathbf{P}_{\tilde{n} \tilde{m}},
\]

are found. The first-order equation associated with the \( e^{-i\omega t} \) term of the coefficients’ response is then given by:

\[
P_C(\omega + \varepsilon - \mathbf{H})\mathbf{C}_u = P_C\{\delta \mathbf{H}_{e^{-i\omega t}}\}\mathbf{C},
\]

\[
\delta \mathbf{H}_{e^{-i\omega t}} = \sum_{j=1}^{Q} \sum_{n_j, m_j=1}^{M_j} \{h_{\tilde{n} \tilde{m}}^j + h_{n_j \delta m_j}^j + \{f_j^\dagger\}_{n_j m_j}\} \mathbf{P}_{n_j m_j}^j + \sum_{\tilde{n}, \tilde{m}} \{\delta W\}_{\tilde{n} \tilde{m}} \mathbf{P}_{\tilde{n} \tilde{m}}.
\]

Similarly, the first-order equation associated with the \( e^{+i\omega t} \) term reads:

\[
P_C^* (\varepsilon - \omega - \mathbf{H}^*)\mathbf{C}_v = P_C^* \{\delta \mathbf{H}_{e^{+i\omega t}}\}^* \mathbf{C}^*,
\]

\[
\{\delta \mathbf{H}_{e^{+i\omega t}}\}^* = \sum_{j=1}^{Q} \sum_{n_j, m_j=1}^{M_j} \{h_{\tilde{n} \tilde{m}}^j + h_{\tilde{n} \delta \tilde{m}}^j + \{f_j^\dagger\}_{m_j n_j}\} \mathbf{P}_{n_j m_j}^j + \sum_{\tilde{n}, \tilde{m}} \{g_j\}_{\tilde{n} \tilde{m}} \mathbf{P}_{\tilde{n} \tilde{m}}.
\]

Compare Eqs. (105) and (106) to the identical-particle case, Eqs. (50) and (51), respectively.
2. Casting the linear-response equations into a matrix form

The quantum object made of the $Q$ coupled degrees-of-freedom responds to the external perturbation as a whole. We thus combine the response amplitudes of all orbitals and expansion coefficients of a given perturbed wavefunction together. The combined response vector:

$$
\begin{pmatrix}
  \mathbf{u}^1 \\
  \vdots \\
  \mathbf{u}^Q \\
  \mathbf{v}^1 \\
  \vdots \\
  \mathbf{v}^Q \\
  \mathbf{C}_u \\
  \mathbf{C}_v
\end{pmatrix}, \quad \mathbf{u}^j = \{|u_{m_j}\rangle\}, \quad \mathbf{v}^j = \{|v_{m_j}\rangle\}, \quad j = 1, \ldots, Q, \ m_j = 1, \ldots, M_j, \quad (107)
$$

is now of length $2(\sum_{j=1}^Q M_j + N_{\text{conf}})$.

Following the strategy taken for identical particles, the final result for the linear-response working matrix equation in the orbital–coefficient response space is given by:

$$
(\mathbf{L} - \omega) \begin{pmatrix}
  \mathbf{u}^1 \\
  \vdots \\
  \mathbf{u}^Q \\
  \mathbf{v}^1 \\
  \vdots \\
  \mathbf{v}^Q \\
  \mathbf{C}_u \\
  \mathbf{C}_v
\end{pmatrix} = \mathbf{R}. \quad (108)
$$

The linear-response matrix $\mathbf{L}$ and the vector $\mathbf{R}$ which collects the various perturbing fields, see Eq. (94), are constructed explicitly below. Analogously to Eq. (54), we term Eq. (108) LR-MCTDH theory.

The solution of the LR-MCTDH linear-response matrix system, Eq. (108), and of the distinguishable-particle Schrödinger equation in linear response requires the eigenvalues $\{\omega_k\}$
and eigenvectors \( \{ R^k \} \) of the linear-response matrix \( \mathbf{L} \):

\[
\mathbf{L} = \omega_k \begin{pmatrix}
\begin{pmatrix} u_{1,k}^1 \\ \vdots \\ u_{Q,k}^1 \\ v_{1,k}^1 \\ \vdots \\ v_{Q,k} \\ C_u^k \\ C_v^k 
\end{pmatrix} \\
\begin{pmatrix} u_{1,k}^1 \\ \vdots \\ u_{Q,k}^1 \\ v_{1,k}^1 \\ \vdots \\ v_{Q,k} \\ C_u^k \\ C_v^k 
\end{pmatrix}
\end{pmatrix} \equiv \omega_k R^k.
\tag{109}
\]

This task is accomplished in Subsec. IV C 3 below.

Now, the matrix form of the linear-response equations of LR-MCTDH, Eq. (108), is assembled as follows. Just like in the identical-particle case, Eq. (56), the linear-response matrix \( \mathbf{L} \) is divided into 4 blocks, \( \mathbf{L}_{oo}, \mathbf{L}_{oc}, \mathbf{L}_{co}, \) and \( \mathbf{L}_{cc} \). The orbital–orbital block \( \mathbf{L}_{oo} \) is further divided into four sub-matrices, like Eq. (57), with the \( \mathbf{L}_{oo}^u \) and \( \mathbf{L}_{oo}^v \) sub-matrices are additionally divided into \( Q \times Q \) rectangular (square on the diagonal) sub-parts each of dimension \( M_j \times M_k \) as follows \((j, k = 1, \ldots, Q)\):

\[
\mathbf{L}_{oo}^{u,jj} = \rho^j \hat{\tau}^j - \mu^j + \Omega^j,
\]

\[
\rho^j = \{ \rho^j_{n_j m_j} \}, \quad \mu^j = \{ \mu^j_{n_j m_j} \}, \quad \Omega^j = \{ \Omega^j_{n_j m_j} \},
\]

\[
\mathbf{L}_{oo}^{u,jk} = \kappa^{1,jk} = \{ \kappa^{1,jk}_{n_j m_k} \} = \left\{ \sum_{\tilde{n}[j], \tilde{m}[k]} \rho_{\tilde{n}\tilde{m}} \tilde{K}_{\tilde{n}[j]\tilde{m}[k]} \right\}, \quad k \neq j,
\]

\[
\mathbf{L}_{oo}^{v,jj} = 0_j,
\]

\[
\mathbf{L}_{oo}^{v,jk} = \kappa^{2,jk} = \{ \kappa^{2,jk}_{n_j m_k} \} = \left\{ \sum_{\tilde{n}[j], \tilde{m}[k]} \rho_{\tilde{n}\tilde{m}} \tilde{K}_{\tilde{n}[j]\tilde{m}[k]} \right\}, \quad k \neq j,
\tag{110}
\]

where \( 0_j \) is a unit matrix of dimension \( M_j \). Note that the diagonal sub-parts \( \mathbf{L}_{oo}^{u,jj} \) of \( \mathbf{L}_{oo}^v \) are zero. This should be contrasted with the respective situation for identical particles, see Eq. (58), where the diagonal sub-matrix \( \mathbf{L}_{oo}^v \) is non-zero. The reason is distinguishability, namely, that in MCTDH there are no two particles associated with the same degree of freedom. Consequently, there are no exchange terms within the same degree-of-freedom of the LR-MCTDH linear-response matrix, compare Eqs. (58) and (110). As can be seen in the latter, there are no exchange operators in neither the \( \mathbf{L}_{oo}^{u,jj} \) nor the \( \mathbf{L}_{oo}^{v,jj} \) diagonal sub-parts.
The orbital–coefficient block $L_{oc}$, like Eq. (60), is comprised of the four sub-matrices where ($j = 1, \ldots, Q, n_j = 1, \ldots, M_j$):

$$L_{\omega_{oc}}^{u,j} = \left\{ C^i \sum_{m_j=1}^{M_j} \left[ \hat{h}^j \rho_{n_jm_j}^{i} + \sum_{\bar{n}[j],\bar{m}[j]} \hat{W}_{\bar{n}[j]\bar{m}[j]} \rho_{\bar{n}\bar{m}} \right] \right\} ,$$

$$L_{\omega_{oc}}^{v,j} = \left\{ C^i \sum_{m_j=1}^{M_j} \left[ \hat{h}^j \rho_{m_jn_j}^{i} + \sum_{\bar{n}[j],\bar{m}[j]} \hat{W}_{\bar{n}[j]\bar{m}[j]} \rho_{\bar{n}\bar{m}} \right] \right\} .$$ (111)

The coefficient–orbital block, like Eq. (62), is comprised of the four sub-matrices with ($j = 1, \ldots, Q, n_j = 1, \ldots, M_j$):

$$L_{\omega_{co}}^{u,j} = \left\{ \sum_{m_j=1}^{M_j} \langle m_j | [\hat{h}^j \rho_{m_jn_j}^{i}] + \sum_{\bar{n}[j],\bar{m}[j]} \hat{W}_{\bar{n}[j]\bar{m}[j]} \rho_{\bar{n}\bar{m}} C \right\} ,$$

$$L_{\omega_{co}}^{v,j} = \left\{ \sum_{m_j=1}^{M_j} \langle m_j | \{ \{ h^j \} \} \rho_{n_jm_j}^{i} + \sum_{\bar{n}[j],\bar{m}[j]} \hat{W}_{\bar{n}[j]\bar{m}[j]} \rho_{\bar{n}\bar{m}} C \right\} .$$ (112)

Furthermore, the relation between the two off-diagonal rectangular blocks of $L$ as in Eq. (64), namely, $(L_{\omega_{oc}}^{u,j})^t = (L_{\omega_{co}}^{v,j})$ and $(L_{\omega_{oc}}^{v,j})^t = (L_{\omega_{co}}^{u,j})$, $j = 1, \ldots, Q$, holds. Finally, the coefficients–coefficients block is given, like Eq. (65), as [102]:

$$L_{cc} = \begin{pmatrix} H - \varepsilon 1_c & 0_c \\ 0_c & -(H^* - \varepsilon 1_c) \end{pmatrix} ,$$ (113)

where $H$ is given in Eq. (103) and, to recall, $1_c$ and $0_c$ are the unit and zero matrices of dimension $N_{\text{conf}}$.

We proceed with the ingredients needed for the linear-response matrix system, as done in Subsec. III E. The combined orbitals–coefficients projector reads:

$$\mathcal{P} = \begin{pmatrix} P_o & 0_{oc} \\ 0_{oc} & P_c \end{pmatrix} , \quad \mathcal{P}_o = \begin{pmatrix} P_o & 0 \\ 0 & P^* \end{pmatrix} , \quad P = \begin{pmatrix} \hat{P}_1 1_e \cdots 0^{1Q} \\ \vdots & \ddots & \vdots \\ 0^{Q1} \cdots \hat{P}_Q 1^Q \end{pmatrix} , \quad \mathcal{P}_c = \begin{pmatrix} P_C & 0_c \\ 0_c & P_C^* \end{pmatrix} ,$$ (114)

where $P$ collects on its diagonal the projectors $\{ \hat{P}_j \}, j = 1, \ldots, Q$ of all degrees-of-freedom, and the dimension of the various unit and zero matrices appearing in Eq. (114) is obvious.
Similarly, the combined orbital–coefficient metric reads:

\[
\mathcal{M} = \begin{pmatrix} \rho_o & 0_{oc} \\ 0_{co} & 1_{2c} \end{pmatrix}, \quad \rho_o = \begin{pmatrix} \rho & 0_o \\ 0_o & \rho^* \end{pmatrix}, \quad \rho = \begin{pmatrix} \rho^1 & \cdots & 0_{o1}^Q \\ \vdots & \ddots & \vdots \\ 0_{o1}^Q & \cdots & \rho^Q \end{pmatrix},
\]

(115)

where \( \rho \) collects on its diagonal the reduced one-body density matrices \( \{ \rho^j \}, j = 1, \ldots, Q \) of all degrees-of-freedom.

To continue, let us collect the perturbing fields in the vector:

\[
\mathcal{M}^{+\frac{1}{2}} \mathcal{R} = \mathcal{M}^{+\frac{1}{2}} + \mathcal{M}^{-\frac{1}{2}} = \begin{pmatrix} -\hat{f}_1^* \phi_1 \\ \vdots \\ -\hat{f}_Q^* \phi_Q \\ \hat{f}_1^* \phi_1 \\ \vdots \\ \hat{f}_Q^* \phi_Q \\ -\sum_{j=1}^Q \sum_{n_j,m_j=1}^{M_j} \{ f_j^\dagger \} n_j m_j \rho_{n_j,m_j}^j C \\ \sum_{j=1}^Q \sum_{n_j,m_j=1}^{M_j} \{ f_j^\dagger \} n_j m_j \rho_{n_j,m_j}^j C^* \end{pmatrix}
\]

(116)

where \( \phi_j = \{ |\phi_{m_j} \rangle \}, j = 1, \ldots, Q, m_j = 1, \ldots, M_j \) are column vectors and \( \Omega^j_g = \{ \Omega^j_{g,n_j,m_j} \} = \left\{ \sum_{\bar{n}[j],\bar{m}[j]} \rho_{\bar{n}\bar{m}} \{ \hat{g}^\dagger \} \bar{n}[j]\bar{m}[j] \right\} \) are square matrices of dimensions \( M_j \times M_j, j = 1, \ldots, Q \). Just as is done in Subsec. [HTP] utilizing the assignments \( \{ \mathcal{P} \mathcal{M}^{+\frac{1}{2}} \mathcal{L} \mathcal{M}^{+\frac{1}{2}} \mathcal{P} \} \Rightarrow \mathcal{L} \), \( \{ \mathcal{P} \mathcal{M}^{+\frac{1}{2}} \mathcal{R} \} \Rightarrow \mathcal{R} \), and \( \{ \mathcal{M}^{+\frac{1}{2}} \left( u^1 \ldots u^Q v^1 \ldots v^Q C_u C_v \right)^\dagger \} \Rightarrow \left( u^1 \ldots u^Q v^1 \ldots v^Q C_u C_v \right) \), we arrive at the linear-response matrix equation [108].

3. Solving the distinguishable-particle Schrödinger equation in linear response

To solve the LR-MCTDH linear-response system, Eq. (108), we have to diagonalize the linear-response matrix \( \mathcal{L} \) and find its excitations energies \( \{ \omega_k \} \) and eigenvectors \( \{ R^k \} \). The analysis of the symmetries and subsequent eigenstates’ resolutions follow exactly the route of the identical-particle linear-response LR-MCTDHX. We capture the essence here for completeness.
We begin with the ‘spin’ matrices \( \Sigma_1 = \begin{pmatrix} \Sigma_1^o & 0_{oc} \\ 0_{co} & \Sigma_1^c \end{pmatrix} \) and \( \Sigma_3 = \begin{pmatrix} \Sigma_3^o & 0_{oc} \\ 0_{co} & \Sigma_3^c \end{pmatrix} \). They have exactly the same appearance and structure as in the identical-particle system, Eqs. (72) and (73), the only difference is the dimension of their orbital entries being \( \sum_{j=1}^Q M_j \). Now, let \( R^k \) be the right eigenvector of \( \mathcal{L} \) with the (real) eigenvalue \( \omega_k \). From the symmetries \( \Sigma_3 \mathcal{L} \Sigma_3 = (\mathcal{L})^\dagger \) and \( \Sigma_1 \mathcal{L} \Sigma_1 = -(\mathcal{L})^* \) we find that \( L^k = \text{sng}^k \Sigma_3 R^k \) [where \( \text{sng}^k \) stands for the sign of the ‘scalar product’ \( \{ (R^k)^\dagger \Sigma_3 R^k \} \)], that \( R^{-k} \equiv \Sigma_1 (R^k)^* \) and \( L^{-k} = -\text{sng}^k \Sigma_3 R^{-k} \), and the orthogonality relations \( (L^k)^\dagger R^{k'} = \text{sng}^k \left[ \sum_{j=1}^Q \left\{ (u^{i,j,k})^\dagger v^{j,k'} - (v^{j,k})^\dagger u^{i,k'} \right\} + (C_{u}^k)^\dagger C_{u}^{k'} - (C_{v}^k)^\dagger C_{v}^{k'} \right] = \delta_{kk'} \) and \( (L^k)^\dagger R^{-k'} = \text{sng}^k \left[ \sum_{j=1}^Q \left\{ (u^{j,k})^\dagger (v^{j,k'})^* - (v^{j,k'})^\dagger (u^{j,k})^* \right\} + (C_{u}^k)^\dagger (C_{v}^{k'})^* - (C_{v}^k)^\dagger (C_{u}^{k'})^* \right] = 0 \). From these, the resolutions of the identify and of the response matrix follow:

\[
1 = \sum_{k=0}^{N_0} \left\{ R^k \mathcal{L}_0^k \right\}^\dagger R^{-k} \left( \mathcal{L}_0^{-k} \right)^\dagger + \sum_{k>N_0} \left\{ R^k \mathcal{L}^k \right\}^\dagger R^{-k} \left( \mathcal{L}^{-k} \right)^\dagger, \quad N_0 = \sum_{j=1}^Q M_j^2;
\]

\[
\mathcal{L} = \sum_{k>N_0} \omega_k \left\{ R^k \mathcal{L}^k - R^{-k} \left( \mathcal{L}^{-k} \right)^\dagger \right\}.
\] (117)

The first group of vectors in Eq. (117) are the zero-mode excitations. Comparison of Eq. (117) for distinguishable degrees-of-freedom to the identical-particle case, Eqs. (77) and (78), shows that the number of zero modes \( N_0 \) is the sum of the squares of \( \{ M_j \} \), \( j = 1, \ldots, Q \), i.e., the number of orbitals used in the ground-state wavefunction for each degree-of-freedom. The second group of vectors are the non-zero-mode excitations. Like above, their summation index, \( k > N_0 \), indicates that we enlist them after the group of zero-mode excitations for which the index of enumeration satisfies \( k \in [0, N_0] \).

Next, expanding the response amplitudes and perturbation with the eigenvectors of \( \mathcal{L} \) and substituting into Eq. (108), we obtain the explicit expression for the system’s response
vector and response weights:

\[
\begin{pmatrix}
  u^1 \\
  \vdots \\
  u^Q \\
  v^1 \\
  \vdots \\
  v^Q \\
  C_u \\
  C_v
\end{pmatrix}
= \sum_{k>N_0} \left[ \frac{\gamma_k}{\omega - \omega_k} R^k + \frac{\gamma_{-k}}{\omega + \omega_k} R^{-k} \right],
\]

\[
\gamma_k = -(L^k)^\dagger R = \text{sgn}^k \left\{ \sum_{j=1}^Q \left( (u^{j,k})^\dagger \left[ (\rho^j)^{+\frac{1}{2}} \hat{f}^j + (\rho^j)^{-\frac{1}{2}} \Omega^{j*}_g \right] \phi_j + \right) \right. \\
\left. + (v^{j,k})^\dagger \left[ (\rho^j)^{+\frac{1}{2}} \hat{f}^j + (\rho^j)^{-\frac{1}{2}} \Omega^{j*}_g \right] \phi_j \right. \\
\left. + (C_u^k)^\dagger \cdot \left[ \sum_{j=1}^{M_j} \sum_{n_j,m_j=1} \{f_j\}_{n_j,m_j} \rho^j_{n_j,m_j} + \sum_{\bar{n},\bar{m}} \{g^j\}_{\bar{n},\bar{m}} \rho_{\bar{n}\bar{m}} \right] C + \right.
\]

\[
\gamma_{-k} = -(L^{-k})^\dagger R = -\text{sgn}^k \left\{ \sum_{j=1}^Q \left( (u^{j,k})^\dagger \left[ (\rho^j)^{+\frac{1}{2}} \hat{f}^j + (\rho^j)^{-\frac{1}{2}} \Omega^{j*}_g \right] \phi_j + \right) \right. \\
\left. + (v^{j,k})^\dagger \left[ (\rho^j)^{+\frac{1}{2}} \hat{f}^j + (\rho^j)^{-\frac{1}{2}} \Omega^{j*}_g \right] \phi_j \right. \\
\left. + (C_v^k)^\dagger \cdot \left[ \sum_{j=1}^{M_j} \sum_{n_j,m_j=1} \{f_j\}_{n_j,m_j} \rho^j_{n_j,m_j} + \sum_{\bar{n},\bar{m}} \{g^j\}_{\bar{n},\bar{m}} \rho_{\bar{n}\bar{m}} \right] C + \right.
\]

\[
+ (C_u^k)^\dagger \cdot \left[ \sum_{j=1}^{M_j} \sum_{n_j,m_j=1} \{f_j\}_{n_j,m_j} \rho^j_{n_j,m_j} + \sum_{\bar{n},\bar{m}} \{g^j\}_{\bar{n},\bar{m}} \rho_{\bar{n}\bar{m}} \right] C^*, \tag{118}
\]

Note that the response weights \{\gamma_k, \gamma_{-k}\} incorporate the response of the entire system of the \(Q\) coupled degrees-of-freedom, via their orbitals and expansion coefficients.

From the solution to the response amplitudes, Eq. (118), and the ansatz for the orbitals and expansion coefficients, Eq. (114), we get the time dependence of the latter in linear

39
\[ \phi_j(r_j, t) \approx \phi_j(r_j) + \delta \phi_j(r_j, t), \quad j = 1, \ldots, Q, \]

\[ \delta \phi_j(r, t) = \sum_{k > N_0} \left\{ \left( \gamma_k \{ \rho^i \} - \frac{1}{2} \{ \nu^{j,k}(r_j) \} e^{-i\omega t} + \gamma_k^* \{ \rho^{i*} \} - \frac{1}{2} \{ \nu^{j,k}(r_j) \}^* e^{+i\omega t} \right) / (\omega - \omega_k) + \right. \]

\[ \left. + \left[ \gamma_{-k} \{ \rho^i \} - \frac{1}{2} \{ \nu^{j,k}(r_j) \}^* e^{-i\omega t} + \gamma_{-k}^* \{ \rho^{i*} \} - \frac{1}{2} \{ \nu^{j,k}(r_j) \} e^{+i\omega t} \right) / (\omega + \omega_k) \right\}, \]

\[ C(t) \approx C + \delta C(t), \]

\[ \delta C(t) = \sum_{k > N_0} \left\{ \left[ \gamma_k C^k_u e^{-i\omega t} + \gamma_k^* \{ C^k_u \}^* e^{+i\omega t} \right] / (\omega - \omega_k) + \right. \]

\[ \left. + \left[ \gamma_{-k} C^k_u e^{-i\omega t} + \gamma_{-k}^* \{ C^k_u \}^* e^{+i\omega t} \right] / (\omega + \omega_k) \right\}, \quad (119) \]

with \( \delta \phi_j(r, t) = \{ \delta \phi_{m_j}(r_j, t), m_j = 1, \ldots, M_j \} \). As before, the orbitals and the expansion coefficients show the largest response at the frequencies \( \{ \pm \omega_k \} \). From Eq. (119) and to complete our task, the time-dependent many-particle wavefunction is given in linear response by:

\[ |\Psi(t)\rangle \approx \sum_n C_n|\bar{n}\rangle + \sum_n \delta C_n(t)|\bar{n}\rangle + \]

\[ \sum_n C_n \left[ \sum_{j=1}^{Q} \sqrt{\langle \delta \phi_{n_j}(r_j, t)|\delta \phi_{n_j}(r_j, t) \rangle} |\bar{n}\rangle \langle \bar{n}| \right], \quad (120) \]

where \( |\bar{n}^{n_j}_{M+1}; t\rangle \) is associated with the (unnormalized) time-dependent \( n_j \)-th response orbital \( \delta \phi_{n_j}(r_j, t) \) of the \( j \)-th degrees-of-freedom. Similarly to above, the response orbitals \( \{ \delta \phi_{n_j}(r_j, t) \} \) need not be orthogonal to each other, unlike their orthogonality with the ground-state orbitals of the \( j \)-th degree-of-freedom which originates from the orbital differential condition \( (12) \). This brings our derivation of LR-MCTDH to a completion.

V. SUMMARY AND CONCLUDING REMARKS

In this work we present a unified representation and view on linear response of systems of interacting particles, let them be identical or distinguishable. The approach is based on the numerically-exact equations of motion of various multiconfigurational time-dependent Hartree (MCTDH) theories. Linearizing MCTDHB and MCTDHF leads to the linear-response theories LR-MCTDHB and LR-MCTDHF for identical bosons and fermions, respectively, whereas linearizing MCTDH for distinguishable degrees-of-freedom leads to the
respective theory LR-MCTDH. Thus, these linear-response theories provide numerically-exact excitation energies and also system’s properties, via the response operators and amplitudes, when numerical convergence is achieved in the calculations.

As a complementary result we introduce an additional projector operator onto the coefficients’ part of MCTDH methods, which leads to a new differential condition for the expansion coefficients within these methods. Together with the famous (orbital) differential condition [28, 29], these two lead to the notion of “fully-projected” MCTDH, and MCTDHB and MCTDHF equations of motion, namely, that the time-dependent multiconfigurational wavepackets evolve in the completely orthogonal space. As a result, the present linear-response theories ensure the response of the orbitals and expansion coefficients to be orthogonal to the stationary (ground-state) wavefunction, because of the appearing orbitals’ and coefficients’ projector operators.

We analyze in particular and explicitly one-body and two-body response operators for identical particles and up to all-system response operators for distinguishable degrees-of-freedom. The resulting working linear-response equations are presented and discussed in detail. The response matrix, which provides the desired excitation energies, does not depend on the special form of the perturbing fields. Consequently, the choice of the perturbing fields can be utilized to study the nature of the respective excited states. Generally, higher-body response operators can give access to more involved excitations. In particular for Bose-Einstein condensates, where experimentally it is a standard practice to alter the particle-particle (two-body) interactions [20, 21], one could naturally expect to access and subsequently analyze new classes of many-body excitations.

The steps to be followed in order to solve the many-body problem in linear response – for identical or distinguishable interacting particles – may be summarized by the following flowchart. First, we calculate the ground state. This is done at a certain level of MCTDHB, MCTDHF, or MCTDH. Second, we construct the linear-response matrix \( \mathbf{L} \). Third, we diagonalize \( \mathbf{L} \) to obtain its eigenvalues – the excitations energies \( \{ \omega_k \} \) – and eigenvectors \( \{ \mathbf{R}^k \} \). Fourth, for a particular choice of the perturbing fields, collected in the response vector \( \mathbf{R} \), the eigenvectors are utilized to compute the response weights \( \{ \gamma_k \} \), which quantify the intensity of the response. Fifth, all these ingredients are combined together to prescribe the time-dependent orbitals and expansion coefficients and hence the many-particle wavefunction \( |\Psi(t)\rangle \) in linear response. The computation of observables and system’s properties
within the linear-response theories presented here, LR-MCTDHB, LR-MCTDHF, and LR-MCTDH, has not been discussed explicitly in the present work, and is deferred to elsewhere.

It is possible to compute low-lying excited states directly by MCTDH methods, see, e.g., Ref. [39]. The resulting wavefunctions are stationary solutions and hence can in principle be utilized as inputs in the above-summarized linear-response procedure. The resulting spectrum and response amplitudes would describe the respective excitations and de-excitations atop these states.

Until now we discussed linear-response theory in bound-state systems. The same formalism can in principle be extended to metastable states. By analytically continuing the response matrix into the complex energy plane, one can also compute metastable excited states of the system and their lifetimes. There are several methods available in the literature to carry out the analytic continuation. One is complex rotation, see, e.g., the reviews [84, 85], and another is by adding a complex absorbing potential to the Hamiltonian of the system, see, e.g., the review [86]. Extending the theory for the linear-response approaches to include metastable states might be more involved than for the reported ones and is not available yet.

The ideas presented here can also be extended to other derived theories emanating from the MCTDH theory, such that for mixtures of identical particles [87, 88], also see Refs. [89, 90]. The separation of the coefficients, which define the reduced density matrices, from the orbitals [79] suggests that other representations for the time-dependent many-body wavepackets would be amenable to linear response in the spirit presented here. For instance, it is possible to envision that propagation theories utilizing other multiconfigurational ansätze – such as, e.g., in multilayer-formulated MCTDH methods [91–94] – would lead to appealing linear-response theories as well. Recent fruitful implementation and applications for trapped identical bosons [71] suggest that much more is to be expected from MCTDH-based linear-response theories.

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Appendix A: The differential condition in MCTDHB and MCTDHF

The differential condition Eq. (19) introduced by the MCTDH developers into the field of multi-dimensional wavepacket propagation [28, 29] is what has made the MCTDH and its daughters such efficient, effective, and practical propagation theories and tools. We have seen that in the context of linear-response theories the differential condition for the orbitals, Eq. (19), together with its analog differential condition for the coefficients, Eq. (26), are what enables us a fully orthogonal response space [also see Eq. (92)]. It is thus instrumental that we recall in this appendix how exactly the differential condition Eq. (19) can be introduced into the MCTDHX (X=B,F) equations of motion, namely, what is the unitary transformation involved.

The equations of motion for the orbitals and expansion coefficients as derived directly from the MCTDHX action–functional are given by (see Refs. [45, 57]):

\[
\hat{P} \sum_{q=1}^{M} [\rho_{pq} (\hat{h} - i \frac{\partial}{\partial t}) + \sum_{s,l=1}^{M} \rho_{plq} \hat{W}_{sl}] |\phi_q \rangle = 0, \quad p = 1, \ldots, M, \\
\mathbf{H}(t) \mathbf{C}(t) = i \frac{\partial \mathbf{C}(t)}{\partial t}, \quad \mathbf{H}_{\vec{n}\vec{n}'}(t) = \langle \vec{n}; t | \hat{H} - i \frac{\partial}{\partial t} | \vec{n}'; t \rangle. \tag{A1}
\]

(A1)

We are looking for such a transformation on the orbitals and expansion coefficients which leaves the many-particle wavefunction invariant,

\[
|\Psi(t)\rangle = \sum_{\vec{n}} C_{\vec{n}}(t) |\vec{n}; t \rangle = \sum_{\vec{n}} \overline{C}_{\vec{n}}(t) |\vec{n}; t \rangle = |\overline{\Psi}(t)\rangle, \tag{A2}
\]

(A2)

but removes the projector \( \hat{P} \) in front of the time derivative. This is readily achieved by the unitary transformation:

\[
|\overline{\phi}_k \rangle = \sum_{p=1}^{M} |\phi_p \rangle U_{pk} \quad \leftrightarrow \quad |\phi_q \rangle = \sum_{j=1}^{M} U_{qj}^{*} |\overline{\phi}_j \rangle, \tag{A3}
\]

(A3)

with

\[
i \hat{U}_{jq} = \sum_{k=1}^{M} -D_{jk} U_{kq}, \quad D_{jk} = i \langle \phi_j | \hat{\phi}_k \rangle, \quad j, k = 1, \ldots, M, \tag{A4}
\]

(A4)

which leads explicitly to the differential condition Eq. (19) for the transformed orbitals:

\[
D_{jk} = i \langle \overline{\phi}_j | \hat{\overline{\phi}}_k \rangle = 0, \quad j, k = 1, \ldots, M. \tag{A5}
\]

(A5)
In Eq. (A5) we employ a shorthand notation where a transformed orbital \( \phi_k \) is denoted by \( \bar{k} \). Substituting Eq. (A3) into the orbital part of Eq. (A1), making use of \( \hat{P} = \bar{\hat{P}} \), of Eqs. (A2;A3) and \( i|\dot{\phi}_q \rangle = \sum_{j=1}^{M}( U_{qj}^{*}|\phi_j \rangle + U_{qj}|\dot{\phi}_j \rangle ) \), \( \bar{\hat{P}}|\dot{\phi}_q \rangle = \sum_{j=1}^{M} U_{qj}^{*}|\dot{\phi}_j \rangle \), and that creation operators transform like Eq. (A3), one readily arrives at the relation:

\[
\hat{P} \sum_{q=1}^{M} \rho_{pq} \hat{h} + \sum_{s,l=1}^{M} \rho_{pql} \hat{W}_{sl} |\phi_q \rangle = \sum_{q=1}^{M} \rho_{pq} |\dot{\phi}_q \rangle, \quad p = 1, \ldots, M.
\]  

(A6)

Eq. (A6) is expressed almost completely with transformed orbitals, that is except of the leftmost creation operator denoted by \( p \). Next, multiplying both sides and summing up by \( \sum_{p=1}^{M} U_{pk} \) (and removing the now superfluous ‘overline’ from all quantities) leads to the equations of motion for the orbitals of MCTDHX, Eq. (17), in their standard, unified form [45, 57].

To show that the same simplification holds for the respective transformed coefficients and their equations of motion, we do so by recalling that these equations are form-invariant (see in this context Ref. [95]). Namely, if \( \mathcal{H}(t)C(t) = i \frac{\partial C(t)}{\partial t} \) are satisfied for the untransformed quantities \( \{\{C_{\vec{n}}(t)\}, \{\phi_k(r, t)\}\} \) then \( \overline{\mathcal{H}(t)C(t)} = i \frac{\partial C(t)}{\partial t} \) are satisfied for the transformed ones \( \{\{\overline{C_{\vec{n}}}(t)\}, \{\overline{\phi}_k(r, t)\}\} \). The proof is straightforward. Equating the variation of the MCTDHX action–functional with respect to the expansion coefficients to zero, the result can be written as follows: \( \langle \vec{n}; t | \hat{H} - i \frac{\partial}{\partial t} |\Psi(t) \rangle = 0, \forall \vec{n}. \) Since, the transformed configurations \( \{\langle \vec{n}; t \rangle\} \) are given as linear combinations of the untransformed configurations \( \{\langle \vec{n}; t \rangle\} \), the operator \( \hat{H} - i \frac{\partial}{\partial t} \) does not depend on the orbitals, and making use of Eq. (A2) we immediately get: \( \langle \vec{n}; t | \hat{H} - i \frac{\partial}{\partial t} |\Psi(t) \rangle = 0, \forall \vec{n}, \) which concludes our proof. To our needs, since the transformed orbitals Eq. (A3) obey the differential condition Eq. (A5), the respective equations of motion for the transformed coefficients (after removing the now superfluous ‘overline’ from all quantities) boil down to the standard equations of motion for the coefficients, Eq. (17).

**Appendix B: Tensor-product representation of vectors and matrix elements in MCTDH**

In this appendix we present the tensor-product representation of quantities in MCTDH, as far as they are needed in the derivation of LR-MCTDH.
The coefficients vector $C$ in MCTDH can be written as follows:

$$C = \sum_{\vec{n}} C_{\vec{n}} \cdot 1^1_{n_1} \otimes \cdots \otimes 1^j_{n_j} \otimes \cdots \otimes 1^Q_{n_Q},$$  \hfill (B1)

where $1^j_{n_j}$ is a column vector of zeros of length $M_j$, except of 1 in the $n_j$-th entry. The time argument is suppressed throughout this appendix without loss of generality. The density-operator matrices for the $j$-th degree-of-freedom can be written as, $n_j, m_j = 1, \ldots, M_j$:

$$\rho^j_{n_j m_j} = (\rho^j_{n_j m_j})^* = 1^1 \otimes \cdots \otimes 1^j_{n_j m_j} \otimes \cdots \otimes 1^Q,$$

$$\rho^j_{m_j n_j} = (\rho^j_{n_j m_j})^t = 1^1 \otimes \cdots \otimes 1^j_{m_j n_j} \otimes \cdots \otimes 1^Q, \quad \text{ (B2)}$$

where $1^k$ is a unit matrix of dimension $M_k$, and $1^j_{m_j n_j}$ is a square matrix of zeros of dimension $M_j$, except of 1 in the $(n_j, m_j)$-th entry. From Eqs. (B1) and (B2) we get the elements of the reduced one-body density matrix Eq. (88) – written in a tensor-product form – as expectation values using the coefficients’ vector:

$$\rho^j_{n_j m_j} = C^\dagger \rho^j_{n_j m_j} C. \quad \text{ (B3)}$$

We can now proceed in a recursive manner (see in this respect Ref. [88]) and assemble higher-order reduced densities. From the two-degrees-of-freedom density operators

$$\rho^{j k}_{n_j m_j n_k m_k} = \rho^j_{n_j m_j} \rho^k_{n_k m_k} = 1^1 \otimes \cdots \otimes 1^j_{n_j m_j} \otimes \cdots \otimes 1^k_{n_k m_k} \otimes \cdots \otimes 1^Q, \quad j \neq k \quad \text{ (B4)}$$

the reduced two-degrees-of-freedom density matrix associated with the $j$-th and $k$-the degrees-of-freedom is given by, $n_j, m_j = 1, \ldots, M_j$ and $n_k, m_k = 1, \ldots, M_k$:

$$\rho^{j k}_{n_j n_k m_j m_k} = C^\dagger \rho^{j k}_{n_j n_k m_j m_k} C. \quad \text{ (B5)}$$

This can be done with higher-order density-operators’ matrices and reduced density matrices up to the all-degrees-of-freedom ones:

$$\rho^{\vec{n} \vec{m}} = \rho^{1}_{n_1 m_1} \cdots \rho^{Q}_{n_Q m_Q} = 1^1_{n_1 m_1} \otimes \cdots \otimes 1^Q_{n_Q m_Q} \quad \text{ (B6)}$$

and

$$\rho^{\vec{n} \vec{m}} = C^\dagger \rho^{\vec{n} \vec{m}} C = C^*_n C_m. \quad \text{ (B7)}$$

From Eq. (B7) we get the elements of the mean-field operators Eq. (89) – written in a tensor-product form – as expectation values using the coefficients’ vector:

$$\hat{\Omega}^j_{n_j m_j} = \sum_{\vec{n}[j], \vec{m}[j]} \hat{W}_{\vec{n}[j] \vec{m}[j]} C^\dagger_{\vec{n} \vec{m}} C, \quad \text { (B8)}$$

45
which enables us to readily perform the variation in Eq. (96).

Finally, the Hamiltonian in the basis of the configurations can be written in an appealing tensor-product form. To this end, consider the column vector of configurations:

$$\mathbf{n}^\dagger = \sum_{\vec{n}} \langle \vec{n} | \cdot 1_{n_1} \otimes \cdots \otimes 1_{n_j} \otimes \cdots \otimes 1_{n_Q}.$$  \hspace{1cm} (B9)

With this notation, the matrix representation of the Hamiltonian Eq. (85) which is given in Eq. (103) takes on the form:

$$\mathbf{H} = \mathbf{n}^\dagger \hat{\mathbf{H}} \mathbf{m},$$  \hspace{1cm} (B10)

which concludes our exposition.

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[96] Otherwise, i.e., for \( \omega = 0 \), the perturbation itself is time-independent, the ansatz for the variation [see second line of Eq. (29)] is also time-independent, and hence Eq. (19) is satisfied automatically. This specific case which is “in-resonance” with the zero-mode excitations formally requires a separate derivation in which the starting point is the response to a time-independent perturbation. However, since we are generally interested in excitations with a non-vanishing frequency (energy), we will not pursue this matter further here.

[97] Utilizing the differential-condition-derived orthogonality relations of the response amplitudes, Eqs. (31) and (33), renders the ‘integration-by-parts’ relations, Eqs. (30) and (32), trivial. Nonetheless, we find it instrumental to explicitly employ also the latter in the derivation of
the linear-response equations in Subsec. III D, also see Ref. 71.

[98] In case the interparticle interactions contain momenta operators, care should be exercised in replacing $\ast$ by $\dagger$ atop an operator like $\delta \hat{W}_{ls}$. Non-local perturbing fields are beyond the scope of the present work.

[99] Strictly speaking, within the Fock-space mapping for identical-particle configurations, Ref. 80, the upper summation $M$ is required only for the creation operators, but it is convenient to leave it also for the annihilation operators.

[100] The shorthand notation for the eigenvectors on the right-hand side of Eq. (55) is also chosen to point out that $\{R^k\}$ are the right eigenvectors of $\mathbf{L}$, and should not be confused with the response vector in the linear-response Eq. (54) denoted by $\mathbf{R}$.

[101] Of course, the metric $\mathbf{M}$ and projector $\mathbf{P}$ matrices within the response matrix $\mathbf{L}$ [see Eq. (71)] comply with these symmetries, explicitly, $\Sigma_1 \mathbf{M} \Sigma_1 = (\mathbf{M})^\ast$, $\Sigma_1 \mathbf{P} \Sigma_1 = (\mathbf{P})^\ast$ and $\Sigma_3 \mathbf{M} \Sigma_3 = (\mathbf{M})^\dagger$, $\Sigma_3 \mathbf{P} \Sigma_3 = (\mathbf{P})^\dagger$.

[102] There is no difference between the $\ast$ atop the identical-particle second-quantized Hamiltonian in Eqs. (51) and (65) and the $\ast$ atop the distinguishable-degrees-of-freedom Hamiltonian-matrix in Eq. (113), because the density-operators’ matrices, see Appendix B, are real-valued matrices.