Multiconfigurational time-dependent Hartree method for mixtures consisting of two types of identical particles

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We specify the formally exact multiconfigurational time-dependent Hartree method originally developed for systems of distinguishable degrees of freedom to mixtures consisting of two types of identical particles. All three cases, Fermi-Fermi, Bose-Bose, and Bose-Fermi mixtures, are treated on an equal footing making explicit use of the reduced one- and two-body density matrices of the mixture. The theory naturally contains as specific cases the versions of the multiconfigurational time-dependent Hartree method for single-species fermions and bosons. Explicit and compact equations of motion are derived and their properties and usage are briefly discussed.

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I. INTRODUCTION

Identical particles fill our quantum world. Protons, neutrons in nuclei; electrons in atoms, molecules and quantum dots; and atoms in quantum fluids and, more recently, in degenerate quantum gases are abundant examples from nuclear, atomic, molecular, and condensed-matter physics. The exploration of quantum dynamics of many-particle systems is of fundamental and practical importance in modern physics [1–5].

The equation governing the dynamics of quantum particles is, in many cases, the well-known time-dependent Schrödinger equation, which can scarcely be solved analytically or exactly. Thus, approximations are a must. The multiconfigurational time-dependent Hartree method (MCTDH) [6,7] is considered at present the most efficient wave-packet propagation approach for distinguishable particles and has successfully and routinely been used for multidimensional dynamical systems consisting of distinguishable degrees of freedom [8–11]. The main idea behind the MCTDH method is to expand the time-dependent many-body wave function of distinguishable particles by *time-dependent* configurations which are optimized according to the Dirac-Frenkel time-dependent variational principle [12,13]. In this way, a much larger effective subspace of the many-particle Hilbert space can be spanned in practice in comparison to multiconfigurational expansions with *stationary* configurations.

The MCTDH approach can treat efficiently dynamical and—using imaginary time-propagation [11]—static properties of few-particle systems. Of course, the system under investigation by MCTDH can consist of identical particles. For instance, we mention that very recently ground- and excited-state properties of weakly to strongly interacting trapped few-boson systems have been studied on a quantitative many-body level by propagating MCTDH in imaginary time [14–16]. Yet, in treating a larger number of identical particles it is essential to use their quantum statistics, Fermi-Dirac or Bose-Einstein, to eliminate the large amount of redundancies of coefficients in the distinguishable-particle multiconfigurational expansion of the MCTDH wave function. Moreover, identical particles commonly interact via two-body interactions, a property which can explicitly be exploited in specifying the MCTDH method for systems of identical particles. Thus, taking explicitly the antisymmetry of the many-fermion wave function to permutations of any two particles into account, the fermionic version of MCTDH—MCTDHF—was independently developed by several groups [17–19]. MCTDHF is currently successfully employed to study many-body dynamics of few-electron systems with or without external fields [20–25]. Not long after, the bosonic version of MCTDH—MCTDHB—was developed in [26,27]. This advancement is, in particular, valuable since very-many bosons can reside in only a small number of orbitals owing to Bose-Einstein statistics. Alternatively speaking, by explicitly exploiting Bose-Einstein statistics it is possible to successfully and quantitatively attack the dynamics of a much larger number of bosons with the MCTDHB theory. As a first application of MCTDHB, the role of excited states in the splitting of a trapped interacting Bose-Einstein condensate by a time-dependent barrier was studied in [26].

The next step was to unify the MCTDH method specified for identical particles, either fermions (MCTDHF) or bosons (MCTDHB), under one formulation [28]. This unified view is not only a compact representation of the problem, rather, it has conceptual and practical advantages. Specifically, the combination of indistinguishability and the two-body interaction leads to an appealing formulation of the propagation equations in terms of the *reduced one- and two-body density matrices* of the system. This opens up further possibilities for approximate self-consistent-like propagation schemes [28]. It is instructive to mention that reduced two-body density matrices is a fruitful and vivid research area including theory and applications in electronic structure of molecules, quantum phase transitions, and ground-state nuclear motion [29–35].

The final step in the hierarchy of many-body propagation

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theories is the specification of the MCTDH approach to *mixtures* of two (or more) kinds of identical particles, which is the purpose of this work. Mixtures appear in many disciplines of physics, for instance: (i) protons and neutrons in nuclei; (ii) electrons and protons in molecules; (iii) fermionic ^{3}He and bosonic ^{4}He as interacting quantum fluids; (iv) electrons and positrons in matter-antimatter systems; and (v) various combinations of fermionic and/or bosonic atoms as degenerate quantum gases. Depending on the quantum statistics of each species, there are three possible mixtures made of two types of identical particles: Fermi-Fermi, Bose-Bose, or Bose-Fermi mixtures. The multiconfigurational time-dependent Hartree method for mixtures (denoted for brevity by MCTDH-*XY*) is derived for the three types of mixtures in a unified manner.

The structure of the paper is as follows. In Sec. II we present the many-body Hamiltonian and multiconfigurational ansatz of a mixture. In Sec. III we develop the working-equations of MCTDH-*XY* in terms of the mixture's reduced one- and two-body density matrices. In Sec. IV we present an extensive discussion and summary. Finally, in the Appendix we collect relevant matrix elements.

II. MANY-BODY HAMILTONIAN AND MULTICONFIGURATIONAL WAVE FUNCTION OF MIXTURES

We consider a mixture of $N=N_A+N_B$ particles; N_A identical particles of type A and N_B identical particles of type B. We would like to treat in a unified manner the three generic cases: (i) both the A and B species are fermions; (ii) both are bosons; or (iii) one of the species is fermions and the second is bosons. For this, we begin with the field operators $\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x})$ and $\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y})$ of the A and B species, respectively, satisfying the usual fermionic (bosonic) anticommutation (commutation) relations,

$$\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x})\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}')\}^\dagger \pm \{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}')\}^\dagger \hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}) = \delta(\mathbf{x}-\mathbf{x}'),$$

$$\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y})\{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}')\}^\dagger \pm \{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}')\}^\dagger \hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}) = \delta(\mathbf{y}-\mathbf{y}'). \tag{1}$$

Of course, the field operators corresponding to different species commute, $[\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}),\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y})]=[\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x})\}^\dagger,\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y})]=0$. The coordinates $\mathbf{x}\equiv\{\mathbf{r},\sigma\}$ and $\mathbf{y}\equiv\{\mathbf{r},\bar{\sigma}\}$ stand for spatial degrees of freedom and (possible) spin. Correspondingly, here and hereafter the following shorthand notation implies: $\delta(\mathbf{x}-\mathbf{x}')=\delta(\mathbf{r}-\mathbf{r}')\delta_{\sigma,\sigma'}$, $\delta(\mathbf{y}-\mathbf{y}')=\delta(\mathbf{r}-\mathbf{r}')\delta_{\bar{\sigma},\bar{\sigma}'}$, $\int d\mathbf{x}\equiv\int d\mathbf{r}\Sigma_\sigma$, and $\int d\mathbf{y}\equiv\int d\mathbf{r}\Sigma_{\bar{\sigma}}$. It is convenient to expand the field operators with two complete sets of orbitals, each comprised of time-dependent orthonormal orbitals,

$$\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x})=\sum_k \hat{a}_k(t)\phi_k(\mathbf{x},t), \quad \hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y})=\sum_{\bar{k}} \hat{b}_{\bar{k}}(t)\psi_{\bar{k}}(\mathbf{y},t). \tag{2}$$

The time-dependent annihilation and corresponding creation operators obey the usual anti-commutation (commutation) relations $\hat{a}_k(t)\hat{a}_q^\dagger(t)\pm\hat{a}_q^\dagger(t)\hat{a}_k(t)=\delta_{kq}$, $\hat{b}_{\bar{k}}(t)\hat{b}_{\bar{q}}^\dagger(t)\pm\hat{b}_{\bar{q}}^\dagger(t)\hat{b}_{\bar{k}}(t)=\delta_{\bar{k}\bar{q}}$ for fermions (bosons) at any time. Of course, annihilation, creation operators belonging to different species commute, $[\hat{a}_k(t),\hat{b}_{\bar{k}}(t)]=[\hat{a}_q^\dagger(t),\hat{b}_{\bar{k}}(t)]=0$, and orbitals belonging to different species need not be orthogonal to one another. Note that the one-particle indices $k,\bar{k}\ldots$ refer to both spatial and spin quantum numbers.

The many-body Hamiltonian of the two-species mixture is conveniently divided into three parts as follows:

$$\hat{H}^{(AB)}=\hat{H}^{(A)}+\hat{H}^{(B)}+\hat{W}^{(AB)},$$

$$\hat{H}^{(A)}=\sum_{k,q} h_{kq}^{(A)}\hat{a}_k^\dagger\hat{a}_q+\frac{1}{2}\sum_{k,s,l,q} W_{ksql}^{(A)}\hat{a}_k^\dagger\hat{a}_s^\dagger\hat{a}_l\hat{a}_q,$$

$$\hat{H}^{(B)}=\sum_{\bar{k},\bar{q}} h_{\bar{k}\bar{q}}^{(B)}\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{q}}+\frac{1}{2}\sum_{\bar{k},\bar{s},\bar{l},\bar{q}} W_{\bar{k}\bar{s}\bar{q}\bar{l}}^{(B)}\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{s}}^\dagger\hat{b}_{\bar{l}}\hat{b}_{\bar{q}},$$

$$\hat{W}^{(AB)}=\sum_{k,\bar{k},q,\bar{q}} W_{k\bar{k}q\bar{q}}^{(AB)}\hat{a}_k^\dagger\hat{a}_q\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{q}}, \tag{3}$$

where $\hat{H}^{(A)},\hat{H}^{(B)}$ are the Hamiltonians of species A,B, and $\hat{W}^{(AB)}$ is the interspecies interaction. Here and hereafter, the dependence on time of quantities is not shown explicitly whenever unambiguous. The matrix elements of the one- and two-body terms of $\hat{H}^{(AB)}$ with respect to the orbitals $\{\phi_k\}$ and $\{\psi_{\bar{k}}\}$ are given by

$$h_{kq}^{(A)}=\int \phi_k^*(\mathbf{x},t)\hat{h}^{(A)}(\mathbf{x})\phi_q(\mathbf{x},t)d\mathbf{x},$$

$$W_{ksql}^{(A)}=\int\int \phi_k^*(\mathbf{x},t)\phi_s^*(\mathbf{x}',t)\hat{W}^{(A)}(\mathbf{x},\mathbf{x}')\phi_q(\mathbf{x},t)\phi_l(\mathbf{x}',t)d\mathbf{x}d\mathbf{x}',$$

$$h_{\bar{k}\bar{q}}^{(B)}=\int \psi_{\bar{k}}^*(\mathbf{y},t)\hat{h}^{(B)}(\mathbf{y})\psi_{\bar{q}}(\mathbf{y},t)d\mathbf{y},$$

$$W_{\bar{k}\bar{s}\bar{q}\bar{l}}^{(B)}=\int\int \psi_{\bar{k}}^*(\mathbf{y},t)\psi_{\bar{s}}^*(\mathbf{y}',t)\hat{W}^{(B)}(\mathbf{y},\mathbf{y}')\psi_{\bar{q}}(\mathbf{y},t)\psi_{\bar{l}}(\mathbf{y}',t)d\mathbf{y}d\mathbf{y}',$$

$$W_{k\bar{k}q\bar{q}}^{(AB)}=\int\int \phi_k^*(\mathbf{x},t)\psi_{\bar{k}}^*(\mathbf{y},t)\hat{W}^{(AB)}(\mathbf{x},\mathbf{y})\phi_q(\mathbf{x},t)\psi_{\bar{q}}(\mathbf{y},t)d\mathbf{x}d\mathbf{y}. \tag{4}$$

Owing to the time-dependent orbitals, the matrix elements (4) are time-dependent quantities even for time-independent one- and two-body operators. We note that the one-body operators $\hat{h}^{(A)}(\mathbf{x})$ and $\hat{h}^{(B)}(\mathbf{y})$ may depend on spin and be time dependent, and that the particle-particle interactions $\hat{W}^{(A)}(\mathbf{x},\mathbf{x}')$, $\hat{W}^{(B)}(\mathbf{y},\mathbf{y}')$, and $\hat{W}^{(AB)}(\mathbf{x},\mathbf{y})$ are general, physical interactions which may also depend on spin and on external, time-dependent fields. The derivation presented here applies for this generic case.

To specify the MCTDH approach to mixtures of identical

particles, the *ansatz* for the many-body wave function $\Psi(t)$ is taken as a linear combination of products of time-dependent configurations as follows:

$$|\Psi(t)\rangle = \sum_{\vec{n},\vec{m}} C_{\vec{n}\vec{m}}(t)|\vec{n};t\rangle \times |\vec{m};t\rangle \equiv \sum_{\vec{n},\vec{m}} C_{\vec{n}\vec{m}}(t)|\vec{n},\vec{m};t\rangle,$$

$$|\vec{n};t\rangle \equiv |n_1,n_2,\ldots,n_M;t\rangle = \frac{1}{\sqrt{n_1!n_2!\cdots n_M!}}(\hat{a}_1^\dagger(t))^{n_1}(\hat{a}_2^\dagger(t))^{n_2}\cdots(\hat{a}_M^\dagger(t))^{n_M}|\mathrm{vac}^{(A)}\rangle,$$

$$|\vec{m};t\rangle = |m_1,m_2,\ldots,m_{\bar{M}};t\rangle = \frac{1}{\sqrt{m_1!m_2!\cdots m_{\bar{M}}!}} \times (\hat{b}_1^\dagger(t))^{m_1}(\hat{b}_2^\dagger(t))^{m_2}\cdots(\hat{b}_{\bar{M}}^\dagger(t))^{m_{\bar{M}}}|\mathrm{vac}^{(B)}\rangle, \quad (5)$$

with the appropriate permutational symmetry. For fermions one chooses the configurations $|\vec{n};t\rangle$, $|\vec{m};t\rangle$ as Slater determinants with time-dependent orbitals and for bosons one employs permanents assembled from time-dependent orbitals. The summation over $\vec{n},\vec{m}$ in Eq. (5) runs over all possible configurations generated by distributing N_A identical particles over the M orbitals $\{\phi_k\}$ and N_B identical particles over the $\bar{M}$ orbitals $\{\psi_{\bar{k}}\}$. It is convenient to collect the individual occupations in the vectors $\vec{n}=(n_1,n_2,\ldots,n_M)$, $\vec{m}=(m_1,m_2,\ldots,m_{\bar{M}})$ where, respectively, $n_1+n_2+\cdots+n_M=N_A$ and $m_1+m_2+\cdots+m_{\bar{M}}=N_B$. For fermions each occupation n_k, $m_{\bar{k}}$ can be either 0 or 1, in accordance with Fermi-Dirac statistics, whereas for bosons n_k, $m_{\bar{k}}$ can take any integer value in the intervals $[0,N_A]$, $[0,N_B]$ as stipulated by Bose-Einstein statistics. $\{C_{\vec{n}\vec{m}}(t)\}$ are the expansion coefficients.

Finally, we obviously have to make use in our propagation theory of the time-derivative $i\frac{\partial}{\partial t}$ operator. The time-derivative $i\frac{\partial}{\partial t}$ acts on the A-species orbitals $\{\phi_k(t)\}$, on the B-species orbitals $\{\psi_{\bar{k}}(t)\}$, and on the expansion coefficients $\{C_{\vec{n}\vec{m}}(t)\}$. Moreover, when acting on the A and B orbitals (subspaces), we find that it is convenient to express the time-derivative operator as a *one-body* operator. These properties are formally expressed by the following relations:

$$i\frac{\partial}{\partial t} \Rightarrow \left(i\frac{\partial}{\partial t}\right)^{(A)} + \left(i\frac{\partial}{\partial t}\right)^{(B)} + \left(i\frac{\partial}{\partial t}\right)^{\{C_{\vec{n}\vec{m}}\}},$$

$$\left(i\frac{\partial}{\partial t}\right)^{(A)} = \sum_{k,q} \hat{a}_k^\dagger \hat{a}_q \left(i\frac{\partial}{\partial t}\right)^{(A)}_{kq}, \qquad \left(i\frac{\partial}{\partial t}\right)^{(A)}_{kq} = i\int \phi_k^*(\mathbf{x},t)\frac{\partial\phi_q(\mathbf{x},t)}{\partial t}d\mathbf{x},$$

$$\left(i\frac{\partial}{\partial t}\right)^{(B)} = \sum_{\bar{k},\bar{q}} \hat{b}_{\bar{k}}^\dagger \hat{b}_{\bar{q}} \left(i\frac{\partial}{\partial t}\right)^{(B)}_{\bar{k}\bar{q}}, \qquad \left(i\frac{\partial}{\partial t}\right)^{(B)}_{\bar{k}\bar{q}} = i\int \psi_{\bar{k}}^*(\mathbf{y},t)\frac{\partial\psi_{\bar{q}}(\mathbf{y},t)}{\partial t}d\mathbf{y}, \quad (6)$$

where the arrow "⇒" indicates that other terms with respect to time differentiation do not appear in the calculation.

III. WORKING EQUATIONS OF THE MULTICONFIGURATIONAL TIME-DEPENDENT HARTREE METHOD FOR MIXTURES (MCTDH-*XY*)

To derive the equations of motion of the multiconfigurational time-dependent Hartree method for mixtures of identical particles we employ the Lagrangian formulation of the time-dependent variational principle [36,37]; also see [27,28]. Practically, we substitute the many-body *ansatz* (5) into the functional action of the time-dependent Schrödinger equation which reads

$$S[\{C_{\vec{n}\vec{m}}(t)\},\{\phi_k(\mathbf{x},t)\},\{\psi_{\bar{k}}(\mathbf{y},t)\}] = \int dt \Bigg\{ \langle\Psi|\hat{\mathcal{H}}^{(AB)}|\Psi\rangle - \sum_{k,j=1}^{M} \mu_{kj}^{(A)}(t)[\langle\phi_k(\mathbf{x},t)|\phi_j(\mathbf{x},t)\rangle - \delta_{kj}] - \sum_{\bar{k},\bar{j}=1}^{\bar{M}} \mu_{\bar{k}\bar{j}}^{(B)}(t)[\langle\psi_{\bar{k}}(\mathbf{y},t)|\psi_{\bar{j}}(\mathbf{y},t)\rangle - \delta_{\bar{k}\bar{j}}] \Bigg\}, \quad (7)$$

where

$$\hat{\mathcal{H}}^{(AB)} = \hat{H}^{(AB)} - i\frac{\partial}{\partial t}. \quad (8)$$

We call $\hat{\mathcal{H}}^{(AB)}$ the many-body Floquet Hamiltonian. The time-dependent Lagrange multipliers $\mu_{kj}^{(A)}(t)$ are introduced to ensure that the time-dependent A-type orbitals $\{\phi_k(\mathbf{x},t)\}$ remain normalized and orthogonal to one another throughout the propagation, and similarly the $\mu_{\bar{k}\bar{j}}^{(B)}(t)$ are for $\{\psi_{\bar{k}}(\mathbf{y},t)\}$. The employment of the Lagrange multipliers allows us to first evaluate the expectation value $\langle\Psi|\hat{\mathcal{H}}^{(AB)}|\Psi\rangle$ and only subsequently perform the variation and require stationarity of the action with respect to its arguments $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$, and $\{C_{\vec{n}\vec{m}}(t)\}$. Two points to remember with respect to the functional action (7) are: the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ and coefficients $\{C_{\vec{n}\vec{m}}(t)\}$ are independent variables of the action (7), and the bra-ket integrations are both in spatial and spin spaces.

To perform the variation of the action (7) with respect to the orbitals we express the expectation value of the many-body Floquet Hamiltonian $\hat{\mathcal{H}}^{(AB)}$ appearing in $S[\{C_{\vec{n}\vec{m}}(t)\},\{\phi_k(\mathbf{x},t)\},\{\psi_{\bar{k}}(\mathbf{y},t)\}]$ in a form which explicitly depends on the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$. Here, we recruit the reduced one- and two-body density matrices of the mixture. Given the normalized wave function $\Psi(t)$, the A and B reduced one-body density matrices read

$$\rho^{(A)}(\mathbf{x}_1|\mathbf{x}_1';t) = \langle\Psi(t)|\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1')\}^\dagger\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1)|\Psi(t)\rangle = \sum_{k,q=1}^{M} \rho_{kq}^{(A)}(t)\phi_k^*(\mathbf{x}_1',t)\phi_q(\mathbf{x}_1,t),$$

$$\rho^{(B)}(\mathbf{y}_1|\mathbf{y}_1';t) = \langle\Psi(t)|\{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1')\}^\dagger\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1)|\Psi(t)\rangle = \sum_{\bar{k},\bar{q}=1}^{\bar{M}} \rho_{\bar{k}\bar{q}}^{(B)}(t)\psi_{\bar{k}}^*(\mathbf{y}_1',t)\psi_{\bar{q}}(\mathbf{y}_1,t). \quad (9)$$

For the matrix elements $\rho_{kq}^{(A)}(t)=\langle\Psi|\hat{a}_k^\dagger\hat{a}_q|\Psi\rangle$ and $\rho_{\bar{k}\bar{q}}^{(B)}(t)=\langle\Psi|\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{q}}|\Psi\rangle$ of the A and B reduced one-body density matrices see the Appendix. It is convenient to collect these matrix elements as $\boldsymbol{\rho}^{(A)}(t)=\{\rho_{kq}^{(A)}(t)\}$, $\boldsymbol{\rho}^{(B)}(t)=\{\rho_{\bar{k}\bar{q}}^{(B)}(t)\}$. Similarly, the A, B and AB reduced two-body density matrices of $\Psi(t)$ are defined by

$$\rho^{(A)}(\mathbf{x}_1,\mathbf{x}_2|\mathbf{x}_1',\mathbf{x}_2';t) = \langle\Psi(t)|\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1')\}^\dagger\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_2')\}^\dagger\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_2)\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1)|\Psi(t)\rangle = \sum_{k,s,l,q=1}^{M} \rho_{kslq}^{(A)}(t)\phi_k^*(\mathbf{x}_1',t)\phi_s^*(\mathbf{x}_2',t)\phi_l(\mathbf{x}_2,t)\phi_q(\mathbf{x}_1,t),$$

$$\rho^{(B)}(\mathbf{y}_1,\mathbf{y}_2|\mathbf{y}_1',\mathbf{y}_2';t) = \langle\Psi(t)|\{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1')\}^\dagger\{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_2')\}^\dagger\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_2)\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1)|\Psi(t)\rangle = \sum_{\bar{k},\bar{s},\bar{l},\bar{q}=1}^{\bar{M}} \rho_{\bar{k}\bar{s}\bar{l}\bar{q}}^{(B)}(t)\psi_{\bar{k}}^*(\mathbf{y}_1',t)\psi_{\bar{s}}^*(\mathbf{y}_2',t)\psi_{\bar{l}}(\mathbf{y}_2,t)\psi_{\bar{q}}(\mathbf{y}_1,t), \quad (10)$$

$$\rho^{(AB)}(\mathbf{x}_1,\mathbf{y}_1|\mathbf{x}_1',\mathbf{y}_1';t) = \langle\Psi(t)|\{\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1')\}^\dagger\hat{\boldsymbol{\Psi}}^{(A)}(\mathbf{x}_1)\{\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1')\}^\dagger \times\hat{\boldsymbol{\Psi}}^{(B)}(\mathbf{y}_1)|\Psi(t)\rangle = \sum_{k,q=1}^{M}\sum_{\bar{k},\bar{q}=1}^{\bar{M}} \rho_{k\bar{k}q\bar{q}}^{(AB)}(t) \times\phi_k^*(\mathbf{x}_1',t)\phi_q(\mathbf{x}_1,t)\psi_{\bar{k}}^*(\mathbf{y}_1',t)\psi_{\bar{q}}(\mathbf{y}_1,t),$$

where the matrix elements $\rho_{kslq}^{(A)}(t)=\langle\Psi|\hat{a}_k^\dagger\hat{a}_s^\dagger\hat{a}_l\hat{a}_q|\Psi\rangle$, $\rho_{\bar{k}\bar{s}\bar{l}\bar{q}}^{(B)}(t)=\langle\Psi|\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{s}}^\dagger\hat{b}_{\bar{l}}\hat{b}_{\bar{q}}|\Psi\rangle$, and $\rho_{k\bar{k}q\bar{q}}^{(AB)}(t)=\langle\Psi|\hat{a}_k^\dagger\hat{a}_q\hat{b}_{\bar{k}}^\dagger\hat{b}_{\bar{q}}|\Psi\rangle$ of the A, B and AB reduced two-body density matrices, respectively, are prescribed in the Appendix.

With these ingredients, the expectation value in the action $S[\{C_{\vec{n}\vec{m}}(t)\},\{\phi_k(\mathbf{x},t)\},\{\psi_{\bar{k}}(\mathbf{y},t)\}]$ can be cast into the compact form

$$\langle\Psi|\hat{\mathcal{H}}^{(AB)}|\Psi\rangle = \sum_{k,q=1}^{M}\rho_{kq}^{(A)}\left[h_{kq}^{(A)} - \left(i\frac{\partial}{\partial t}\right)_{kq}^{(A)}\right] + \frac{1}{2}\sum_{k,s,l,q=1}^{M}\rho_{kslq}^{(A)}W_{ksql}^{(A)} + \sum_{\bar{k},\bar{q}=1}^{\bar{M}}\rho_{\bar{k}\bar{q}}^{(B)}\left[h_{\bar{k}\bar{q}}^{(B)} - \left(i\frac{\partial}{\partial t}\right)_{\bar{k}\bar{q}}^{(B)}\right] + \frac{1}{2}\sum_{\bar{k},\bar{s},\bar{l},\bar{q}=1}^{\bar{M}}\rho_{\bar{k}\bar{s}\bar{l}\bar{q}}^{(B)}W_{\bar{k}\bar{s}\bar{q}\bar{l}}^{(B)} + \sum_{k,q=1}^{M}\sum_{\bar{k},\bar{q}=1}^{\bar{M}}\rho_{kk\bar{q}\bar{q}}^{(AB)}W_{kk\bar{q}\bar{q}}^{(AB)} - i\sum_{\vec{n}\vec{m}}C_{\vec{n}\vec{m}}^*\frac{\partial C_{\vec{n}\vec{m}}}{\partial t}. \quad (11)$$

Expression (11) is appealing because it depends on the reduced one- and two-body density matrices $\rho^{(A)}(\mathbf{x}_1|\mathbf{x}_1';t)$, $\rho^{(B)}(\mathbf{y}_1|\mathbf{y}_1';t)$ and $\rho^{(A)}(\mathbf{x}_1,\mathbf{x}_2|\mathbf{x}_1',\mathbf{x}_2';t)$, $\rho^{(B)}(\mathbf{y}_1,\mathbf{y}_2|\mathbf{y}_1',\mathbf{y}_2';t)$, $\rho^{(AB)}(\mathbf{x}_1,\mathbf{y}_1|\mathbf{x}_1',\mathbf{y}_1';t)$. Side by side, the only explicit dependence of Eq. (11) on the orbitals $\{\phi_k(\mathbf{x},t)\}$ and $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ is grouped into the matrix elements $h_{kq}^{(A)}$, $\left(i\frac{\partial}{\partial t}\right)_{kq}^{(A)}$, $W_{ksql}^{(A)}$; $h_{\bar{k}\bar{q}}^{(B)}$, $\left(i\frac{\partial}{\partial t}\right)_{\bar{k}\bar{q}}^{(B)}$, $W_{\bar{k}\bar{s}\bar{q}\bar{l}}^{(B)}$; and $W_{k\bar{k}q\bar{q}}^{(AB)}$ whereas the elements of the mixture's reduced one- and two-body density matrices *do not* depend explicitly on the orbitals.

We can now perform the variation of the functional action (7) with respect to the orbitals. Using the fact that the sets of orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ comprise orthonormal functions we eliminate, respectively, the Lagrange multipliers $\mu_{kj}^{(A)}(t)$, $\mu_{\bar{k}\bar{j}}^{(B)}(t)$ and arrive at the following equations of motion for the time-dependent orbitals, $j=1,\ldots,M$, $\bar{j}=1,\ldots,\bar{M}$:

$$\hat{\mathbf{P}}^{(A)}i|\dot{\phi}_j\rangle = \hat{\mathbf{P}}^{(A)}\left[\hat{h}^{(A)}|\phi_j\rangle + \sum_{k,q=1}^{M}\{\boldsymbol{\rho}^{(A)}(t)\}_{jk}^{-1} \times\left\{\sum_{s,l=1}^{M}\rho_{kslq}^{(A)}\hat{W}_{sl}^{(A)} + \sum_{\bar{k},\bar{q}=1}^{\bar{M}}\rho_{k\bar{k}q\bar{q}}^{(AB)}\hat{W}_{\bar{k}\bar{q}}^{(AB)}\right\}|\phi_q\rangle\right],$$

$$\hat{\mathbf{P}}^{(B)}i|\dot{\psi}_{\bar{j}}\rangle = \hat{\mathbf{P}}^{(B)}\left[\hat{h}^{(B)}|\psi_{\bar{j}}\rangle + \sum_{\bar{k},\bar{q}=1}^{\bar{M}}\{\boldsymbol{\rho}^{(B)}(t)\}_{\bar{j}\bar{k}}^{-1} \times\left\{\sum_{\bar{s},\bar{l}=1}^{\bar{M}}\rho_{\bar{k}\bar{s}\bar{l}\bar{q}}^{(B)}\hat{W}_{\bar{s}\bar{l}}^{(B)} + \sum_{k,q=1}^{M}\rho_{k\bar{k}q\bar{q}}^{(AB)}\hat{W}_{kq}^{(BA)}\right\}|\psi_{\bar{q}}\rangle\right]. \quad (12)$$

Here,

$$\hat{\mathbf{P}}^{(A)} = 1 - \sum_{j'=1}^{M}|\phi_{j'}\rangle\langle\phi_{j'}|, \quad \hat{\mathbf{P}}^{(B)} = 1 - \sum_{\bar{j}'=1}^{\bar{M}}|\psi_{\bar{j}'}\rangle\langle\psi_{\bar{j}'}| \quad (13)$$

are, respectively, projection operators onto the subspaces orthogonal to these spanned by the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$,

$$\hat{W}_{sl}^{(A)}(\mathbf{x},t) = \int\phi_s^*(\mathbf{x}',t)\hat{W}^{(A)}(\mathbf{x},\mathbf{x}')\phi_l(\mathbf{x}',t)d\mathbf{x}',$$

$$\hat{W}_{\bar{s}\bar{l}}^{(B)}(\mathbf{y},t) = \int\psi_{\bar{s}}^*(\mathbf{y}',t)\hat{W}^{(B)}(\mathbf{y},\mathbf{y}')\psi_{\bar{l}}(\mathbf{y}',t)d\mathbf{y}',$$

$$\hat{W}_{\bar{k}\bar{q}}^{(AB)}(\mathbf{x},t) = \int\psi_{\bar{k}}^*(\mathbf{y},t)\hat{W}^{(AB)}(\mathbf{x},\mathbf{y})\psi_{\bar{q}}(\mathbf{y},t)d\mathbf{y},$$

$$\hat{W}_{kq}^{(BA)}(\mathbf{y},t) = \int \phi_k^*(\mathbf{x},t)\hat{W}^{(AB)}(\mathbf{x},\mathbf{y})\phi_q(\mathbf{x},t)d\mathbf{x} \tag{14}$$

are (for spin-independent interactions) *local* time-dependent potentials, and $\dot{\phi}_j \equiv \frac{\partial \phi_j}{\partial t}$, $\dot{\psi}_{\bar{j}} \equiv \frac{\partial \psi_{\bar{j}}}{\partial t}$. Examining Eq. (12) we see that eliminating the Lagrange multipliers $\mu_{kj}^{(A)}(t)$, $\mu_{\bar{k}\bar{j}}^{(B)}(t)$ has emerged as the projection operators $\hat{\mathbf{P}}^{(A)}$, $\hat{\mathbf{P}}^{(B)}$. These projection operators appear both on the left- and right-hand sides of Eq. (12), making Eq. (12) a cumbersome coupled system of integrodifferential nonlinear equations.

To simplify the equations of motion (12) we note that the many-body wave function (5) is invariant under independent unitary transformations of the A and B orbitals, compensated by the "reverse" transformations of the coefficients $\{C_{\vec{n}\vec{m}}(t)\}$. Fortunately, there exists *one* specific unitary transformation, which guarantees without introducing further constraints that the conditions [6,7,28]

$$\langle \phi_k(\mathbf{x},t)|\dot{\phi}_q(\mathbf{x},t)\rangle = 0, \quad k,q = 1,\ldots,M,$$

$$\langle \psi_{\bar{k}}(\mathbf{y},t)|\dot{\psi}_{\bar{q}}(\mathbf{y},t)\rangle = 0, \quad \bar{k},\bar{q} = 1,\ldots,\bar{M} \tag{15}$$

are satisfied at any point in time. Obviously, when conditions (15) are satisfied at any time, the A, and independently, the B orbitals remain orthonormal functions at any time. This representation simplifies considerably the equations of motion (12) which now read, $j=1,\ldots,M$, $\bar{j}=1,\ldots,\bar{M}$,

$$\begin{aligned} i|\dot{\phi}_j\rangle &= \hat{\mathbf{P}}^{(A)}\Bigg[\hat{h}^{(A)}|\phi_j\rangle + \sum_{k,q=1}^{M}\{\boldsymbol{\rho}^{(A)}(t)\}_{jk}^{-1} \\ &\times\Bigg\{\sum_{s,l=1}^{M}\rho_{kslq}^{(A)}\hat{W}_{sl}^{(A)} + \sum_{\bar{k},\bar{q}=1}^{\bar{M}}\rho_{k\bar{k}q\bar{q}}^{(AB)}\hat{W}_{\bar{k}\bar{q}}^{(AB)}\Bigg\}|\phi_q\rangle\Bigg], \\ i|\dot{\psi}_{\bar{j}}\rangle &= \hat{\mathbf{P}}^{(B)}\Bigg[\hat{h}^{(B)}|\psi_{\bar{j}}\rangle + \sum_{\bar{k},\bar{q}=1}^{\bar{M}}\{\boldsymbol{\rho}^{(B)}(t)\}_{\bar{j}\bar{k}}^{-1} \\ &\times\Bigg\{\sum_{\bar{s},\bar{l}=1}^{\bar{M}}\rho_{\bar{k}\bar{s}\bar{l}\bar{q}}^{(B)}\hat{W}_{\bar{s}\bar{l}}^{(B)} + \sum_{k,q=1}^{M}\rho_{k\bar{k}q\bar{q}}^{(AB)}\hat{W}_{kq}^{(BA)}\Bigg\}|\psi_{\bar{q}}\rangle\Bigg]. \end{aligned} \tag{16}$$

The projection operators $\hat{\mathbf{P}}^{(A)}$, $\hat{\mathbf{P}}^{(B)}$ remaining on the right-hand side of Eq. (16) make it clear that conditions (15) are satisfied at any point in time throughout the propagation of the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$. In practice, the meaning of the conditions $\langle \phi_k(\mathbf{x},t)|\dot{\phi}_q(\mathbf{x},t)\rangle = 0$ and $\langle \psi_{\bar{k}}(\mathbf{y},t)|\dot{\psi}_{\bar{q}}(\mathbf{y},t)\rangle = 0$ is that the temporal changes of the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ are always orthogonal to $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ themselves. This property originally introduced by the MCTDH developers [6,7] generally makes the time propagation of Eq. (16) robust and stable and can thus be exploited to maintain accurate propagation results at lower computational costs.

To complete the derivation, we perform the variation of Eq. (7) with respect to the coefficients $\{C_{\vec{n}\vec{m}}(t)\}$, which is easily done after expressing the expectation value of $\hat{\mathcal{H}}^{(AB)}$ in a form which explicitly depends on the $\{C_{\vec{n}\vec{m}}(t)\}$,

$$\begin{aligned} &\langle\Psi|\hat{\mathcal{H}}^{(AB)}|\Psi\rangle \\ &= \sum_{\vec{n},\vec{m}} C_{\vec{n},\vec{m}}^*\Bigg[\sum_{\vec{n}',\vec{m}'}\langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(AB)}|\vec{n}',\vec{m}';t\rangle C_{\vec{n}',\vec{m}'} - i\frac{\partial C_{\vec{n},\vec{m}}}{\partial t}\Bigg]. \end{aligned} \tag{17}$$

The following result then straightforwardly emerges:

$$\boldsymbol{\mathcal{H}}^{(AB)}(t)\mathbf{C}(t) = i\frac{\partial \mathbf{C}(t)}{\partial t},$$

$$\mathcal{H}_{\vec{n}\vec{m},\vec{n}'\vec{m}'}^{(AB)}(t) = \langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(AB)}|\vec{n}',\vec{m}';t\rangle, \tag{18}$$

where the vector $\mathbf{C}(t)$ collects the coefficients $\{C_{\vec{n}\vec{m}}(t)\}$. The matrix elements of the many-body Floquet operator $\hat{\mathcal{H}}^{(AB)}$ with respect to two general configurations $|\vec{n},\vec{m};t\rangle$ and $|\vec{n}',\vec{m}';t\rangle$ are prescribed in a *unified* manner in the Appendix for the *three* possible types of mixtures: Fermi-Fermi, Bose-Bose, or Bose-Fermi. Finally, making use of conditions (15) we obtain the familiar MCTDH form of the equations of motion for the propagation of the coefficients,

$$\mathbf{H}^{(AB)}(t)\mathbf{C}(t) = i\frac{\partial \mathbf{C}(t)}{\partial t}, \quad H_{\vec{n}\vec{m},\vec{n}'\vec{m}'}^{(AB)}(t) = \langle\vec{n},\vec{m};t|\hat{H}^{(AB)}|\vec{n}'\vec{m}';t\rangle. \tag{19}$$

The coupled equations of motion (12) for the orbitals $\{\phi_j(\mathbf{x},t)\}$, $\{\psi_{\bar{j}}(\mathbf{y},t)\}$ and Eq. (18) for the expansion coefficients $\{C_{\vec{n},\vec{m}}(t)\}$, or, respectively, Eqs. (16) and (19) constitute the multiconfigurational time-dependent Hartree method for mixtures consisting of two types of identical particles (MCTDH-XY); let them be Fermi-Fermi, Bose-Bose, or Bose-Fermi mixtures.

IV. DISCUSSION AND SUMMARY

A many-body propagation theory for mixtures consisting of two types of identical particles (MCTDH-XY) has been derived by specifying the multiconfigurational time-dependent Hartree method (MCTDH) originally developed for distinguishable particles. The recent successful applications of the single-species propagation approaches for fermionic (MCTDHF) and bosonic (MCTDHB) systems, in combination with the employment of reduced one- and two-body density matrices, solicit the application of the present theory for studying the many-body quantum dynamics of mixtures.

A. General aspects of MCTDH-XY

In the MCTDH-XY theory, the ansatz for the many-particle wave function is taken as a linear combination of all possible *time-dependent* configurations made by distributing the A-species particles over M *time-dependent* orbitals $\{\phi_k(\mathbf{x},t)\}$ and distributing the B-species particles over $\bar{M}$ *time-dependent* orbitals $\{\psi_{\bar{k}}(\mathbf{y},t)\}$. The evolution of the

many-body wave function is then determined by utilizing a standard time-dependent variational principle within the Lagrangian formulation. Performing the variation, we arrive at two sets of coupled equations of motion, one for the orbitals and one for the expansion coefficients. The first, Eq. (12) or Eq. (16), is for the evolution of the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$, which assemble the configurations $|\vec{n},\vec{m};t\rangle$. The second, Eq. (18) or Eq. (19), is for the expansion coefficients $\{C_{\vec{n}\vec{m}}(t)\}$ in the multiconfigurational wave function $|\Psi(t)\rangle$. In the most general case, the particles in the mixture have also spin degrees of freedom. This is fully accounted for in the MCTDH-*XY* theory by employing orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$ that depend on spatial and spin coordinates, i.e., spin orbitals.

There are three possible propagation schemes—for Fermi-Fermi (MCTDH-FF), for Bose-Bose (MCTDH-BB), and for Bose-Fermi (MCTDH-BF) mixtures—and the formulation we employ treats them all in a unified manner. Indeed, the only difference in particle statistics appears in our derivation explicitly only in the first of the above equations, namely, Eq. (1) for the field operators $\hat{\Psi}^{(A)}(\mathbf{x})$ and $\hat{\Psi}^{(B)}(\mathbf{y})$. Implicitly, the differences in particle statistics translate themselves into the form of the reduced one-body $\rho^{(A)}(\mathbf{x}_1|\mathbf{x}_1';t)$, $\rho^{(B)}(\mathbf{y}_1|\mathbf{y}_1';t)$ and two-body $\rho^{(A)}(\mathbf{x}_1,\mathbf{x}_2|\mathbf{x}_1',\mathbf{x}_2';t)$, $\rho^{(B)}(\mathbf{y}_1,\mathbf{y}_2|\mathbf{y}_1',\mathbf{y}_2';t)$, $\rho^{(AB)}(\mathbf{x}_1,\mathbf{y}_1|\mathbf{x}_1',\mathbf{y}_1';t)$ density matrices and the matrix representation of the many-body Floquet Hamiltonian $\hat{\mathcal{H}}^{(AB)}$ evaluated between two general configurations $|\vec{n},\vec{m};t\rangle$ and $|\vec{n}',\vec{m}';t\rangle$. We prescribe in the Appendix these matrix elements for the three types of mixtures in a unified manner.

The equations of motion of MCTDH-*XY*, Eqs. (12) and (18), or Eqs. (16) and (19) become an exact representation of the time-dependent many-particle Schrödinger equation in the limit where the number of orbitals $M,\bar{M}$ goes to infinity. In practice, one has of course to limit $M,\bar{M}$ where the employment of time-dependent orbitals, which is at the heart and success of MCTDH, MCTDHF, and MCTDHB, is of great advantage. Still, even with time-dependent orbitals the actual size of the Hilbert subspace rapidly increases with the number of particles N_A,N_B and the number of orbitals $M,\bar{M}$ employed. Thus, for large systems and for strong interparticle interactions it is instructive to devise strategies for further approximations atop the multiconfigurational expansion (5), which utilizes complete Hilbert subspaces, i.e., all configurations resulting by distributing N_A,N_B particles over $M,\bar{M}$ orbitals are explicitly taken into account. We mention here in brief that, owing to the structure of the MCTDH-*XY* equations, there are two natural strategies to devise approximations: one is based on restricting the number of configurations taken into account in the many-body wave function (5); the other is based on writing equations of motion for the reduced one- and two-body density matrices (9) and (10) themselves and approximating them. Developing these strategies for the quantum dynamics of mixtures is a theme beyond this work.

Two final remarks. In the absence of interspecies interactions the MCTDH-*XY* equations boil down to two sets of single-species equations, either for fermions (MCTDHF [17–19,28]) and/or for bosons (MCTDHB [26–28]), depending on the particles in the mixture. The extension of MCTDH-*XY* to mixtures consisting of three or more types of identical particles is straightforwardly performed along the lines presented here.

B. Mixtures of polarized particles

For a general many-body Hamiltonian $\hat{H}^{(AB)}$, the forces between the particles may be spin dependent. In such a case the dynamics of the mixture would involve changes in both spatial and spin degrees of freedom. We remind that the equations of motion (16) and (19) [or Eqs. (12) and (18); hereafter not referred to for brevity] of the MCTDH-*XY* are fully equipped to describe the time evolution of the generic case.

Next, consider a mixture of polarized particles, say spin-half fermions with spin projection $S_z=+\frac{1}{2}$ [species A] and spin-one bosons with spin projection $\bar{S}_z=-1$ [species B] prepared initially at $t=0$. Furthermore, suppose that the many-body Hamiltonian $\hat{H}^{(AB)}$ preserves the spin projection of each species, i.e., there are no spin-orbit and spin-spin coupling terms, nor external fields which lead to spin flips. We can of course use equations of motion (16) and (19) to propagate the dynamics of this system and obtain, as anticipated due to $\hat{H}^{(AB)}$, that no other spin projections are populated in time.

As symmetry is usually helpful in simplifying the treatment of quantum systems, see, e.g., [2], it can help us here as well. Specifically, in such a case there is no need to work with the (full) spin orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$. Instead, we can work with the respective components $\{\phi_k(\mathbf{r},\sigma=+\frac{1}{2},t)\}$, $\{\psi_{\bar{k}}(\mathbf{r},\bar{\sigma}=-1,t)\}$ of the spin orbitals only. In practice, the orbitals $\{\phi_k(\mathbf{r},\sigma=+\frac{1}{2},t)\}$, $\{\psi_{\bar{k}}(\mathbf{r},\bar{\sigma}=-1,t)\}$ satisfy the same MCTDH-*XY* equations of motion (16) and (19) when excluding therein summation over the spin coordinates Σ_σ, $\Sigma_{\bar{\sigma}}$ and leaving the spatial integrations $\int d\mathbf{r}$ only. In turn, Eqs. (16) and (19) are now to be utilized with the one-body $h_{kq}^{(A)}$, $h_{\bar{k}\bar{q}}^{(B)}$ and two-body $W_{ksql}^{(A)}$, $W_{\bar{k}\bar{s}\bar{q}\bar{l}}^{(B)}$, $W_{k\bar{k}q\bar{q}}^{(AB)}$ matrix elements, the projection operators $\hat{\mathbf{P}}^{(A)}$, $\hat{\mathbf{P}}^{(B)}$, and the time-dependent matrix elements of $\hat{H}^{(AB)}$ between two general configurations which are all to be evaluated with orbitals, rather than with spin orbitals, thus saving unnecessary effort.

Next, we examine a *single-species* system comprised of particles with spin S, either fermions or bosons. We assume spin-independent interactions. We can treat the evolution of the single-species system in two ways. In the first, we recruit the single-species MCTDHF or MCTDHB approaches for fermions or bosons. In this case, the orbitals in use are, in general, spin orbitals. It is well known, however, that identical particles with different spin projections can be treated as distinguishable particles [38]. Thus, we also can treat single-species spin-S particles with the multiconfigurational time-dependent Hartree approach for mixtures. Taking a system comprised of electrons as an example, the spin-up and spin-down electrons would house now the A and B species in the "mixture." Where is the advantage? The orbitals of each species are purely spatial functions now, which saves the same

type of overheads described in the previous example.

Combining the above two examples, we finally look at different mixtures comprising two kinds of identical particles with or without spin, such as (i) mixtures of spinless bosons; (ii) mixtures of spinless bosons and unpolarized or polarized spin-half fermions; (iii) mixtures of two kinds of spin-half fermions; and (iv) mixtures of spin S bosons and spin $\bar{S}$ bosons; etc. Spin-independent interactions are assumed. We may now time propagate these systems in two ways: (1) using the MCTDH-*XY* equations of motion (16) and (19) with spin orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$, or (2) counting the *total* number of spin projections in the mixture and treating the system as a "new" mixture with, in general $2(S+\bar{S}+1)$ "species." This would transform the spin-orbitals to purely spatial functions thus simplifying the equations of motion. The propagation equations of such "multicomponent" mixtures are straightforwardly derived along the lines presented in this work.

C. Stationary many-body states of mixtures

The presently developed multiconfigurational time-dependent propagation theory can be applied via imaginary-time propagation to compute ground- and excited-state properties of mixtures. In the spirit of Refs. [11,39], one may also arrive at a multiconfigurational self-consistent theory for stationary properties of mixtures. Setting $t\rightarrow -it$, the left-hand sides of Eqs. (12) and (16) decay to zero in time. Then, by translating back from the projection operators $\hat{\mathbf{P}}^{(A)}$, $\hat{\mathbf{P}}^{(B)}$ to the Lagrange multipliers $\mu_{kj}^{(A)}$, $\mu_{\bar{k}\bar{j}}^{(B)}$, we arrive at the multiconfigurational self-consistent (time-independent) working equations for the orbitals as follows:

$$\sum_{j=1}^{M}\left[\rho_{kj}^{(A)}\hat{h}^{(A)}+\sum_{s,l=1}^{M}\rho_{kslj}^{(A)}\hat{W}_{sl}^{(A)}+\sum_{\bar{k},\bar{q}=1}^{\bar{M}}\rho_{\bar{k}kj\bar{q}}^{(AB)}\hat{W}_{\bar{k}\bar{q}}^{(AB)}\right]|\phi_j\rangle = \sum_{j=1}^{M}\mu_{kj}^{(A)}|\phi_j\rangle,\ k=1,\ldots,M,$$

$$\sum_{\bar{j}=1}^{\bar{M}}\left[\rho_{\bar{k}\bar{j}}^{(B)}\hat{h}^{(B)}+\sum_{\bar{s},\bar{l}=1}^{\bar{M}}\rho_{\bar{k}\bar{s}\bar{l}\bar{j}}^{(B)}\hat{W}_{\bar{s}\bar{l}}^{(B)}+\sum_{k,q=1}^{M}\rho_{k\bar{k}q\bar{j}}^{(AB)}\hat{W}_{kq}^{(BA)}\right]|\psi_{\bar{j}}\rangle = \sum_{\bar{j}=1}^{\bar{M}}\mu_{\bar{k}\bar{j}}^{(B)}|\psi_{\bar{j}}\rangle,\ \bar{k}=1,\ldots,\bar{M}. \tag{20}$$

Making use of the fact that the matrices of Lagrange multipliers $\{\mu_{kj}^{(A)}\}$, $\{\mu_{\bar{k}\bar{j}}^{(B)}\}$ are Hermitian (for stationary states), one may work in a representation of Eq. (20) in which these matrices are taken to be diagonal. The reason, as mentioned above, is that the multiconfigurational wave function [see Eq. (5)] is invariant under independent unitary transformations of the A and B orbitals, which can be exploited to independently diagonalize the matrices of the Lagrange multipliers $\{\mu_{kj}^{(A)}\}$, $\{\mu_{\bar{k}\bar{j}}^{(B)}\}$. Last, setting $t\rightarrow -it$ and on similar grounds, Eqs. (18) and (19) reduce to the (time-independent) eigenvalue problem

$$\mathbf{H}^{(AB)}\mathbf{C}=\varepsilon\mathbf{C}, \tag{21}$$

where ε is the eigenenergy of the mixture.

Equations (20) and (21) constitute a multiconfigurational self-consistent theory for mixtures. This theory generalizes the available multiconfigurational self-consistent-field theories for fermions [40,41] and for bosons [39].

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APPENDIX: EVALUATING MATRIX ELEMENTS OF OPERATORS AND REDUCED DENSITY MATRICES WITH MULTICONFIGURATIONAL WAVEFUNCTIONS OF MIXTURES

The equations of motion of the MCTDH-*XY* theory employ two types of matrix elements: matrix elements of the mixture's reduced density matrices with respect to the orbitals $\{\phi_k(\mathbf{x},t)\}$, $\{\psi_{\bar{k}}(\mathbf{y},t)\}$, and matrix elements of the many-body Floquet Hamiltonian with respect to the configurations $|\vec{n},\vec{m};t\rangle$. In this appendix we prescribe the evaluation of these matrix elements.

The matrix elements themselves depend on the particles' statistics. As we saw in the main text, there are three cases: Fermi-Fermi (MCTDH-FF), Bose-Bose (MCTDH-BB), and Bose-Fermi (MCTDH-BF). We can treat the matrix elements in all three cases together, in the spirit of unifying the single-species (MCTDHF and MCTDHB) matrix elements done in Ref. [28]. In Ref. [28] we prescribed the elements of the reduced one- and two-body density matrices for single-species multiconfigurational fermionic [42] and bosonic [39] wave functions, $|\Psi(t)\rangle=\Sigma_{\vec{n}}C_{\vec{n}}(t)|n_1,n_2,\ldots,n_M;t\rangle$, in a *unified* manner. Aiming at an economical account, we present and discuss here only the *differences and additions* to the single-species (fermions or bosons) matrix elements unified in [28].

We begin with the matrix elements of the A and B reduced density matrices, Eqs. (9) and (10). Owing to the many-body wave function being expressed as the sum of products of single-species configurations, the mixture's reduced A and B density matrices take on a form resembling the single-species reduced density matrices as follows:

$$\rho_{kq}^{(A)}(t)=\langle\Psi|\hat{a}_k^\dagger\hat{a}_q|\Psi\rangle=\sum_{\vec{n},\vec{n}'}\left\{\sum_{\vec{m}}C_{\vec{n},\vec{m}}^{*}C_{\vec{n}',\vec{m}}\right\}\langle\vec{n};t|\hat{a}_k^\dagger\hat{a}_q|\vec{n}';t\rangle,$$

$$\rho^{(B)}_{\bar{k}\bar{q}}(t) = \langle\Psi|\hat{b}^\dagger_{\bar{k}}\hat{b}_{\bar{q}}|\Psi\rangle = \sum_{\vec{m},\vec{m}'}\left\{\sum_{\vec{n}} C^*_{\vec{n},\vec{m}}C_{\vec{n},\vec{m}'}\right\}\langle\vec{m};t|\hat{b}^\dagger_{\bar{k}}\hat{b}_{\bar{q}}|\vec{m}';t\rangle,$$

$$\rho^{(A)}_{kslq}(t) = \langle\Psi|\hat{a}^\dagger_k\hat{a}^\dagger_s\hat{a}_l\hat{a}_q|\Psi\rangle = \sum_{\vec{n},\vec{n}'}\left\{\sum_{\vec{m}} C^*_{\vec{n},\vec{m}}C_{\vec{n}',\vec{m}}\right\}\langle\vec{n};t|\hat{a}^\dagger_k\hat{a}^\dagger_s\hat{a}_l\hat{a}_q|\vec{n}';t\rangle,$$

$$\rho^{(B)}_{\bar{k}\bar{s}\bar{l}\bar{q}}(t) = \langle\Psi|\hat{b}^\dagger_{\bar{k}}\hat{b}^\dagger_{\bar{s}}\hat{b}_{\bar{l}}\hat{b}_{\bar{q}}|\Psi\rangle = \sum_{\vec{m},\vec{m}'}\left\{\sum_{\vec{n}} C^*_{\vec{n},\vec{m}}C_{\vec{n},\vec{m}'}\right\}\langle\vec{m};t|\hat{b}^\dagger_{\bar{k}}\hat{b}^\dagger_{\bar{s}}\hat{b}_{\bar{l}}\hat{b}_{\bar{q}}|\vec{m}';t\rangle. \tag{A1}$$

In other words, the matrix element of the mixture's A and B reduced density matrices can be directly read from the matrix elements of the single-species ones [28] by writing instead of the single-species expansion coefficients $C_{\vec{n}}$ the respective $C_{\vec{n},\vec{m}}$ coefficients of the mixture and adding summation over the second-species occupations—$\vec{m}$ ($\vec{n}$) for the A (B) species.

To evaluate the AB reduced two-body density matrix (and later on the matrix elements of the interspecies interaction term $\hat{W}^{(AB)}$) we need a shorthand notation for configurations in mixtures of identical particles. Let the reference configuration be denoted by $|\vec{n},\vec{m};t\rangle = |n_1,\ldots,n_k,\ldots,n_q,\ldots,n_M:m_1,\ldots,m_{\bar{k}},\ldots,m_{\bar{q}},\ldots,m_{\bar{M}};t\rangle$. Then, the configuration denoted by $|\vec{n}^q_k,\vec{m};t\rangle = |n_1,\ldots,n_k-1,\ldots,n_q+1,\ldots,n_M:m_1,\ldots,m_{\bar{k}},\ldots,m_{\bar{q}},\ldots,m_{\bar{M}};t\rangle$ differs from $|\vec{n},\vec{m};t\rangle$ by a transfer of one A particle from the kth to the qth orbital of the A species; $|\vec{n},\vec{m}^{\bar{q}}_{\bar{k}};t\rangle = |n_1,\ldots,n_k,\ldots,n_q,\ldots,n_M:m_1,\ldots,m_{\bar{k}}-1,\ldots,m_{\bar{q}}+1,\ldots,m_{\bar{M}};t\rangle$ differs from $|\vec{n},\vec{m};t\rangle$ by a transfer of one B particle from the $\bar{k}$th to the $\bar{q}$th orbital of the B species; and $|\vec{n}^q_k,\vec{m}^{\bar{q}}_{\bar{k}};t\rangle = |n_1,\ldots,n_k-1,\ldots,n_q+1,\ldots,n_M:m_1,\ldots,m_{\bar{k}}-1,\ldots,m_{\bar{q}}+1,\ldots,m_{\bar{M}};t\rangle$ differs from $|\vec{n},\vec{m};t\rangle$ by a transfer of one A and one B particles, an A particle from the kth to the qth orbital of the A species, and a B particle from the $\bar{k}$th to the $\bar{q}$th orbital of the B species. Note that we employ a nomenclature in which the *same* ordering of the orbitals $\phi_1,\phi_2,\ldots,\phi_M$ and $\psi_1,\psi_2,\ldots,\psi_{\bar{M}}$ as in Eq. (5) is kept in *all* configurations. Next, to represent in a unified way the restriction on the maximal number of particles per orbitals for fermionic and bosonic species we employ the following notation. Let p_A,p_B be the maximal number of particles per orbitals of the A,B species. Obviously $p_A=1$, $p_B=1$ for fermionic atoms and $p_A=N_A$, $p_B=N_B$ for bosonic atoms. We define $[n]_A=n$ if $0\leqslant n\leqslant p_A$ and $[n]_A=0$ otherwise, and similarly $[m]_B=m$ if $0\leqslant m\leqslant p_B$ and $[m]_B=0$ otherwise. Finally, we define the "distance" between the kth and qth entries, $k<q$, in the A species configuration $|\vec{n};t\rangle$ as $d^{kq}_{\vec{n}}=\Sigma^q_{l=k+1}n_l$, $n_l\in\vec{n}$. Similarly, we define the distance between the $\bar{k}$th and $\bar{q}$th entries, $\bar{k}<\bar{q}$, in the B species configuration $|\vec{m};t\rangle$ as $d^{\bar{k}\bar{q}}_{\vec{m}}=\Sigma^{\bar{q}}_{\bar{l}=\bar{k}+1}m_{\bar{l}}$, $m_{\bar{l}}\in\vec{m}$. The quantities $d^{kq}_{\vec{n}},d^{\bar{k}\bar{q}}_{\vec{m}}$ are needed for fermionic-species matrix elements due to the anticommutation relation between fermionic creation operators. In the equations below the upper sign refers to fermionic species and the lower sign to bosonic species.

With these conventions, the matrix elements of the AB reduce two-body density matrix $\rho^{(AB)}(\mathbf{x}_1,\mathbf{y}_1|\mathbf{x}'_1,\mathbf{y}'_1;t)$ given the multiconfigurational wave function $\Sigma_{\vec{n},\vec{m}}C_{\vec{n},\vec{m}}(t)|\vec{n},\vec{m};t\rangle$ are

$$\rho_{kk\bar{k}\bar{k}} = \sum_{\vec{n},\vec{m}} C^*_{\vec{n},\vec{m}}C_{\vec{n},\vec{m}}n_k m_{\bar{k}},$$

$$\rho_{kk\bar{k}\bar{q}} = \sum_{\vec{n},\vec{m}} C^*_{\vec{n},\vec{m}}C_{\vec{n}^q_k,\vec{m}}\sqrt{n_k[n_q+1]_A}m_{\bar{k}}(\mp1)^{\{d^{kq}_{\vec{n}}\}},\ k<q$$

$$\rho_{k\bar{k}k\bar{q}} = \sum_{\vec{n},\vec{m}} C^*_{\vec{n},\vec{m}}C_{\vec{n},\vec{m}^{\bar{q}}_{\bar{k}}}\sqrt{m_{\bar{k}}[m_{\bar{q}}+1]_B}n_k(\mp1)^{\{d^{\bar{k}\bar{q}}_{\vec{m}}\}},\ \bar{k}<\bar{q}$$

$$\rho_{k\bar{k}q\bar{q}} = \sum_{\vec{n},\vec{m}} C^*_{\vec{n},\vec{m}}C_{\vec{n}^q_k,\vec{m}^{\bar{q}}_{\bar{k}}}\sqrt{n_k[n_q+1]_A}\sqrt{m_{\bar{k}}[m_{\bar{q}}+1]_B} \times\begin{cases}(\mp1)^{\{d^{kq}_{\vec{n}}+d^{\bar{k}\bar{q}}_{\vec{m}}\}}, & k<q,\bar{k}<\bar{q}\\ (\mp1)^{\{d^{kq}_{\vec{n}}+d^{\bar{q}\bar{k}}_{\vec{m}}-1\}}, & k<q,\bar{q}<\bar{k}.\end{cases} \tag{A2}$$

All other nonvanishing matrix elements can be computed due to the hermicity of the density $\rho^*_{k\bar{k}q\bar{q}}=\rho_{q\bar{q}k\bar{k}}$.

We now move to the matrix elements of the many-body Floquet Hamiltonian [see Eqs. (3) and (8)] with respect to the configurations $|\vec{n},\vec{m};t\rangle$. In Ref. [28] we presented in a *unified* manner Slater-Condon rules for evaluating matrix elements with determinants [40] and their bosonic analog for evaluating matrix elements with permanents [39]. We proceed with this line here, expressing the matrix elements of the many-body Floquet Hamiltonian (8) of Fermi-Fermi, Bose-Bose, and Bose-Fermi mixtures in a unified manner. Owing to the structure of the many-body Hamiltonian $\hat{H}^{(AB)}$, and combining Eqs. (3), (6), and (8), we get for the many-body Floquet Hamiltonian,

$$\langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(AB)}|\vec{n}'\vec{m}';t\rangle = \langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(A)}+\hat{\mathcal{H}}^{(B)}+\hat{W}^{(AB)}|\vec{n}'\vec{m}';t\rangle, \tag{A3}$$

where $\hat{\mathcal{H}}^{(A)}=\hat{H}^{(A)}-(i\frac{\partial}{\partial t})^{(A)}$ and $\hat{\mathcal{H}}^{(B)}=\hat{H}^{(B)}-(i\frac{\partial}{\partial t})^{(B)}$. Consequently, we obviously have for the first two, single-species terms,

$$\langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(A)}|\vec{n}'\vec{m}';t\rangle = \delta_{\vec{m},\vec{m}'}\langle\vec{n};t|\hat{\mathcal{H}}^{(A)}|\vec{n}';t\rangle,$$

$$\langle\vec{n},\vec{m};t|\hat{\mathcal{H}}^{(B)}|\vec{n}'\vec{m}';t\rangle = \delta_{\vec{n},\vec{n}'}\langle\vec{m};t|\hat{\mathcal{H}}^{(B)}|\vec{m}';t\rangle. \tag{A4}$$

These matrix elements are obviously read from the single-species ones [28].

Finally, with the above conventions and the matrix elements $W^{(AB)}_{kk\bar{q}\bar{q}}$ of the interspecies interaction potential $\hat{W}^{(AB)}(\mathbf{x},\mathbf{y})$ with respect to the orbitals [see Eq. (4)], the nonvanishing matrix elements of the interspecies two-body (second-quantized) operator $\hat{W}^{(AB)}$ [see Eq. (3)] with respect to the configurations $|\vec{n},\vec{m};t\rangle$ follow from

$$\langle \vec{n},\vec{m};t|\hat{W}^{(AB)}|\vec{n},\vec{m};t\rangle = \sum_{l=1}^{M}\sum_{\bar{l}=1}^{\bar{M}} n_l m_{\bar{l}} W^{(AB)}_{\bar{l}\bar{l}ll},$$

$$\langle \vec{n},\vec{m};t|\hat{W}^{(AB)}|\vec{n}_k^q,\vec{m};t\rangle = \sqrt{n_k[n_q+1]_A}\sum_{\bar{l}=1}^{\bar{M}} m_{\bar{l}} W^{(AB)}_{k\bar{l}q\bar{l}}(\mp 1)^{\{d_{\vec{n}}^{kq}\}},$$

$$k<q,$$

$$\langle \vec{n},\vec{m};t|\hat{W}^{(AB)}|\vec{n},\vec{m}_{\bar{k}}^{\bar{q}};t\rangle = \sqrt{m_{\bar{k}}[m_{\bar{q}}+1]_B}\sum_{l=1}^{M} n_l W^{(AB)}_{l\bar{k}l\bar{q}}(\mp 1)^{\{d_{\vec{m}}^{\bar{k}\bar{q}}\}},$$

$$\bar{k}<\bar{q},$$

$$\langle \vec{n},\vec{m};t|\hat{W}^{(AB)}|\vec{n}_k^q,\vec{m}_{\bar{k}}^{\bar{q}};t\rangle = \sqrt{n_k[n_q+1]_A}\sqrt{m_{\bar{k}}[m_{\bar{q}}+1]_B} W^{(AB)}_{k\bar{k}q\bar{q}} \begin{cases} (\mp 1)^{\{d_{\vec{n}}^{kq}+d_{\vec{m}}^{\bar{k}\bar{q}}\}}, & k<q,\bar{k}<\bar{q}, \\ (\mp 1)^{\{d_{\vec{n}}^{kq}+d_{\vec{m}}^{\bar{q}\bar{k}}-1\}}, & k<q,\bar{q}<\bar{k}, \end{cases} \tag{A5}$$

and the fact that $\hat{W}^{(AB)}$ is self-adjoint, $\langle \vec{n},\vec{m};t|\hat{W}^{(AB)}|\vec{n}',\vec{m}';t\rangle = \langle \vec{n}',\vec{m}';t|\hat{W}^{(AB)}|\vec{n},\vec{m};t\rangle^*$.

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