Coupled-cluster calculations for valence systems around ^{16}O

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We study the ground and low-lying excited states of ^{15}O, ^{17}O, ^{15}N, and ^{17}F using modern two-body nucleon-nucleon interactions and the suitably designed variants of the *ab initio* equation-of-motion coupled-cluster theory aimed at an accurate description of systems with valence particles and holes. A number of properties of ^{15}O, ^{17}O, ^{15}N, and ^{17}F, including ways the energies of ground and excited states of valence systems around ^{16}O change as functions of the number of nucleons, are correctly reproduced by the equation-of-motion coupled-cluster calculations performed in up to eight major-oscillator shells. Certain disagreements with experiment are in part because of the degrees of freedom such as three-body interactions not accounted for in our effective two-body Hamiltonians. In particular, the calculated binding energies of ^{15}O/^{15}N and ^{17}O/^{17}F enable us to rationalize the discrepancy between the experimental and recently published [Phys. Rev. Lett. **94**, 212501 (2005)] equation-of-motion coupled-cluster excitation energies for the $J^{\pi} = 3^{-}$ state of ^{16}O. Our calculations demonstrate the feasibility of the equation-of-motion coupled-cluster methods to deal with valence systems around closed-shell nuclei and to provide results for systems beyond $A = 16$.

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I. INTRODUCTION

The way shell closures and single-particle energies evolve as functions of the number of nucleons is presently one of the greatest challenges to our understanding of the basic features of nuclei. The properties of single-particle energies and states with a strong quasiparticle content along an isotopic chain are moreover expected to be strongly influenced by the nuclear spin-orbit force. The latter can be retraced to contributions from both two-body and three-body models of the nuclear forces (see, for example, Refs. [1,2]). A fully microscopic *ab initio* description of masses, shell closures, excited states, and single-particle energies in terms of the underlying nuclear forces is an unresolved problem in nuclear physics that awaits a satisfactory and computationally tractable solution.

For light nuclei with mass numbers $A \sim 12$, both Green's function Monte Carlo methods [3] and large-scale no-core shell-model calculations [4,5] provide almost converged benchmarks for selected two- and three-body Hamiltonians, where typically the models for the two-body nucleon-nucleon interactions reproduce the available scattering data, whereas the three-body interaction models are normally fit to reproduce the binding energies of selected nuclei. The agreement with experimental data for many light nuclei is quite reasonable in these calculations. Unfortunately, for medium-mass and heavier nuclei the dimensionality of the corresponding many-particle problem becomes intractable by the Green's function Monte Carlo methods and *ab initio* no-core shell-model techniques, and one typically has to resort to a simplified shell-model description within a smaller space, the so-called model space. To solve the corresponding many-body Schrödinger equation, one needs then to derive effective two- and/or three-body interactions for the chosen small model space. Many-body perturbation theory is normally employed to derive effective interactions [6], but unless these interactions are fitted to reproduce selected properties of nuclei [7,8], one cannot correctly recover the experimentally derived single-particle and excitation energies and shell closures (see, for example, Ref. [9]).

Two key points make it imperative to investigate new theoretical methods that will allow for an accurate description of closed- as well as open-shell nuclei with $A \gg 12$. First, present and proposed nuclear structure research facilities will open significant territory into regions of medium-mass and heavier nuclei, where the majority of the studied nuclei will be open-shell systems and where many of the nuclei produced in experiment will be unstable or short lived. Second, existing shell-model and Green's function Monte Carlo techniques have prohibitive computer costs that scale factorially or exponentially with the system size. In addition to an increased dimensionality, one needs to account for the fact that many of the medium-mass and heavier nuclei can be weakly bound and couple to resonant states. Moreover, to examine new nuclei that have not been discovered or studied before, one may not be able to rely on fitting the effective Hamiltonians to the experimental data for the known nuclei, as has been traditionally done for many years. Microscopic *ab initio* methods, in which nuclear properties are obtained from the underlying nucleon-nucleon interactions, will become increasingly important as the new information about the medium-mass and heavier nuclei is obtained in various experiments. In addition to these practical aspects, *ab initio* calculations of nuclear properties, including, for example, the way the binding and excitation energies change as a function of the number of nucleons around closed-shell nuclei, may provide important new insights into our understanding of nuclear forces.

Clearly, if we wish to extend *ab initio* methods to nuclei with $A \gg 12$, we have to consider alternatives to the existing Green's function Monte Carlo and no-core shell-model techniques. In this work, we focus on coupled-cluster theory [10–13], which is a promising candidate for the development of practical methods for fully microscopic *ab initio* studies of nuclei in the $A \gg 12$ mass region. As has been demonstrated in numerous quantum chemistry applications (see, e.g., Refs. [14–24] for selected reviews), coupled-cluster methods are capable of providing a precise description of many-particle correlation effects at relatively low computer costs, when compared to shell-model or configuration interaction techniques aimed at similar accuracies. Based on the remarkable success of coupled-cluster methods in chemistry and molecular physics, where one has to obtain a highly accurate description of many-electron correlation effects, we believe that the field of nuclear physics may significantly benefit from the vast experience in the development of accurate and computationally efficient coupled-cluster approximations and algorithms developed by quantum chemists.

Although historically coupled-cluster theory originated in nuclear physics [10,11], its applications to the nuclear many-body problem have been relatively rare (see, e.g., Refs. [25–27] and references therein), particularly when compared to quantum chemistry. For many years, part of the problem has been an inadequate understanding of nucleon-nucleon interactions and lack of adequate computer resources in the 1970s and 1980s. This situation has changed only in the past few years. The successful construction of realistic nucleon-nucleon potentials (cf., e.g., Refs. [28–31]) and spectacular improvements in computer technology have led to renewed interest in applying coupled-cluster methods in *ab initio* nuclear physics calculations. In particular, using bare interactions, Mihaila and Heisenberg performed impressive coupled-cluster calculations for the binding energy and the electron scattering form factor of ^{16}O [32–35]. Because of their use of bare interactions, the convergence with increasing model-space size was quite slow. We have taken an alternative route and combined a few basic coupled-cluster techniques with the renormalized form of the Hamiltonian to determine ground and selected excited states of ^{4}He and ^{16}O [36–41], demonstrating promising results when compared with the results of the exact shell-model diagonalization in the same model space [37] and, at least for some properties, with the experimental data [36,38–41]. In particular, in our most recent study of the ground and excited states of ^{16}O [38], we obtained fully converged results that are very close to those obtained with more expensive large-scale no-core shell-model calculations of Navrátil and collaborators [42]. This has been possible thanks to the use of the diagram factorization techniques [43], which lead to almost perfectly vectorized [24,44,45] and highly scalable parallel [36] computer codes, enabling routine calculations for systems in the $A \sim 20$ region with large single-particle basis sets, including seven or even eight major oscillator shells (336 and 480 single-particle states, respectively).

Our initial coupled-cluster calculations [36–41] focused on closed-shell nuclei; however, one long-term objective of our research program is to study open-shell nuclei with one or more valence nucleons. We would like, for example, to examine how the binding and excitation energies vary with the number of nucleons in valence systems around closed-shell nuclei. This is particularly interesting when we examine the $A = 15$ and $A = 17$ nuclei around ^{16}O. For example, the splittings between the $(3/2)^-_1$ and $(1/2)^-_1$ states in ^{15}O and ^{15}N and the splittings between the $(3/2)^+_1$ and $(5/2)^+_1$ states in ^{17}O and ^{17}F should arise from the nuclear spin-orbit force, which may or may not be affected by three-nucleon interactions. One would like to examine such issues by comparing the results of *ab initio* calculations employing two-body interactions with the experimental energy spacings. This requires an appropriate extension of the usual single-reference ground-state coupled-cluster theory [10–13] to ground and excited states of valence systems around closed-shell nuclei.

In this article we examine, for the first time, the applicability of two quantum-chemistry-inspired coupled-cluster approaches, referred to as the particle-attached (PA) (in chemistry, electron-attached or EA [46–50]) and particle-removed (PR) (in chemistry, ionized or IP [16,46,50–56]) equation-of-motion coupled-cluster (EOMCC) methods [46,57,58], in the converged calculations of the binding and excitation energies of $A = 15$ (^{15}O, ^{15}N) and $A = 17$ (^{17}O, ^{17}F) nuclei. For these calculations, we use modern nucleon-nucleon interactions derived from the effective-field theory [59,60], such as N^{3}LO [31], and their slightly older phenomenological counterparts, including the charge-dependent Bonn interaction model (CD-Bonn) [29] and the V_{18} model of the Argonne group [28]. In the PA- and PR-EOMCC methods, one calculates ground and excited states of the $(A + 1)$- and $(A - 1)$-particle systems by diagonalizing the similarity-transformed Hamiltonian of the coupled-cluster theory, resulting from the ground-state calculations for the A-particle closed-shell nucleus, in the relevant $(A + 1)$- and $(A - 1)$-particle subspaces of the Fock space. As shown in this article, the PA- and PR-EOMCC approaches provide us with practical computational techniques for potentially accurate *ab initio* studies of valence systems around the closed-shell nuclei. Application of these methods may provide several important insights into the effects of the underlying nucleon-nucleon interactions on the calculated properties of such systems. We also provide in this article several details of the PA-EOMCC, PR-EOMCC, and underlying ground-state coupled-cluster calculations, including the factorized forms of the relevant amplitude equations.

This article is divided into four sections. In Sec. II, we present our formalism for deriving an effective two-body Hamiltonian for coupled-cluster calculations, which takes into account short-range nucleon-nucleon correlations, and present the details of the PA-EOMCC and PR-EOMCC theories that enable us to deal with valence systems around closed-shell nuclei within the framework of the single-reference coupled-cluster formalism. The results of PA-EOMCC and PR-EOMCC calculations for the ^{15}O, ^{17}O, ^{15}N, and ^{17}F nuclei are discussed in Sec. III and the conclusions and perspectives are outlined in Sec. IV. The factorized forms of the PA-EOMCC and PR-EOMCC equations for the $(A \pm 1)$-particle systems and the corresponding ground-state coupled-cluster equations, exploited in this work, are given in the Appendix.

II. COUPLED-CLUSTER EQUATIONS FOR VALENCE SYSTEMS

Our theoretical considerations start with the introduction of an appropriate two-body effective interaction for the large-scale coupled-cluster calculations. The relevant information used to renormalize the bare nucleon-nucleon Hamiltonians and to generate the final effective Hamiltonians corrected for the center-of-mass contaminations are discussed in Sec. II A. We then provide in Sec. II B the most essential information about the underlying closed-shell coupled-cluster calculations that precede the PA- and PR-EOMCC steps. In Sec. II C, we discuss the most essential details of the PA- and PR-EOMCC calculations for the ground and excited states of the $(A+1)$- and $(A-1)$-particle valence systems around the closed-shell A-particle nucleus. In Sec. II D we briefly discuss further computational details of the PA- and PR-EOMCC calculations.

A. Effective two-body interaction for coupled-cluster calculations

In this work, we concentrate on results obtained from the N^{3}LO [31] nucleon-nucleon interaction, and we also give some results for the CD-Bonn [29] and V_{18} [28] potentials. The Coulomb interaction was included perturbatively in all of our calculations (to distinguish between ^{15}O/^{17}O and ^{15}N/^{17}F). This lack of renormalization of the Coulomb leads, however, to a weak dependence on the oscillator energy when we compare the binding energies of these systems. To remove the hard-core part of the interaction and enable realistic calculations in manageable model spaces, we follow the procedure exploited in our earlier work [36–41]. Thus, we renormalize the Hamiltonian through a no-core G-matrix procedure, described in considerable detail in Refs. [6,36]. The no-core G-matrix approach introduces a starting-energy (ω) dependence in the effective two-body matrix elements $G(\omega)$ defining the renormalized two-body interactions (obtained by analyzing the exactly solvable proton-proton, proton-neutron, and neutron-neutron two-body problems), but much of the ω dependence can be eliminated through the use of the Bethe-Brandow-Petschek theorem [61] and the appropriate summation of the class of folded diagrams to infinite order at a given starting energy (see Refs. [6,36] for further information). For nuclei like ^{16}O, the dependence on the chosen starting energy ω is weak. In our calculations the starting energies are defined by the energy of the hole states and are typically in the range $\omega \in [-50, -10]$ MeV. This introduces an uncertainty of 0.1–0.2 MeV per particle for the binding energies.

After renormalizing bare interactions with the G-matrix approach, our effective Hamiltonian is given by the formula

$$H_{\rm eff}(\omega) = H_0 + G(\omega), \tag{1}$$

where H_0 is the translationally invariant kinetic energy

$$H_0 = \sum_{i=1}^{A} \frac{\mathbf{p_i}^2}{2m} - \frac{\mathbf{P}^2}{2mA}, \tag{2}$$

and m is the nucleon mass. Here the center-of-mass momentum for an A-body system is $\mathbf{P} = \sum_{\mathbf{i}=1}^{\mathbf{A}} \mathbf{p_i}$. The momenta $\mathbf{p_i}$ are the single-particle momenta. In our actual calculations we rewrite Eq. (2) as

$$H_0 = \left(1 - \frac{1}{A}\right) \sum_{i=1}^{A} \frac{\mathbf{p_i}^2}{2m} - \sum_{i<j}^{A} \frac{\mathbf{p_i}\cdot\mathbf{p_j}}{mA}. \tag{3}$$

The last term of this equation is computed separately as an expectation value, after we have optimized the total center-of-mass contribution, as discussed in the next steps.

To complete the process of preparing the Hamiltonian for coupled-cluster calculations, we correct the renormalized Hamiltonian $H_{\rm eff}(\omega)$, Eq. (1), resulting from exploiting the no-core G-matrix procedure, for center-of-mass contaminations using the expression

$$\begin{aligned} H \equiv H(\omega, \beta_{\rm CoM}) &= H_{\rm eff}(\omega) + \beta_{\rm CoM} H_{\rm CoM} \\ &= z^{\beta}_{\alpha} a^{\alpha} a_{\beta} + \tfrac{1}{4} v^{\gamma\delta}_{\alpha\beta} a^{\alpha} a^{\beta} a_{\delta} a_{\gamma}, \end{aligned} \tag{4}$$

where $z^{\beta}_{\alpha} = \langle\alpha|z|\beta\rangle$ and $v^{\gamma\delta}_{\alpha\beta} = \langle\alpha\beta|v|\gamma\delta\rangle - \langle\alpha\beta|v|\delta\gamma\rangle$ are the relevant one- and two-body matrix elements in a single-particle basis set $\{|\alpha\rangle\}$ and a^{α} (a_{α}) are the usual creation (annihilation) operators. We use the Einstein summation convention over repeated upper and lower indices.

In our coupled-cluster derivations, we use the normal-ordered form of the Hamiltonian, H_N, relative to the A-particle Fermi vacuum reference state $|\Phi\rangle$,

$$H_N = H - \langle\Phi|H|\Phi\rangle = f^{\beta}_{\alpha} N[a^{\alpha} a_{\beta}] + \tfrac{1}{4} v^{\gamma\delta}_{\alpha\beta} N[a^{\alpha} a^{\beta} a_{\delta} a_{\gamma}], \tag{5}$$

where $f^{\beta}_{\alpha} \equiv \langle\alpha|f|\beta\rangle = z^{\beta}_{\alpha} + v^{\beta i}_{\alpha i}$ are matrix elements of the Fock matrix and $N[\dots]$ designates the normal product.

The center-of-mass term is given by

$$H_{\rm CoM} = \frac{\mathbf{P}^2}{2MA} + \frac{1}{2} mA\Omega^2 \mathbf{R}^2 - \frac{3}{2}\hbar\Omega, \tag{6}$$

where $\mathbf{R} = \left(\sum_{\mathbf{i}=1,\mathbf{A}} \mathbf{r_i}\right)/\mathbf{A}$. The term $H_{\rm CoM}$ can be rewritten as a one-body harmonic potential and a two-body term that depends on both the relative and center-of-mass coordinates of the two interacting particles. The parameter $\beta_{\rm CoM}$ is chosen such that the expectation value of the center-of-mass Hamiltonian $H_{\rm CoM}$ with the ground-state coupled-cluster wave function, $\langle H_{\rm CoM}\rangle$, obtained for the $\beta_{\rm CoM}$-dependent Hamiltonian H, Eq. (4), is 0.0 MeV [62]. This can be done by relying on the Hellmann-Feynman theorem and calculating $\langle H_{\rm CoM}\rangle$ as the first derivative of the coupled-cluster energy with respect to $\beta_{\rm CoM}$. As pointed out in our earlier articles [38,40,41], one of the advantages of this procedure is the ease of separation of intrinsic and center-of-mass contaminated states by analyzing the dependence of coupled-cluster energies on $\beta_{\rm CoM}$. As shown in Refs. [40,41], the physical states obtained in coupled-cluster calculations are independent of $\beta_{\rm CoM}$, whereas the center-of-mass contaminated states show a strong, nearly linear dependence of excitation energies on $\beta_{\rm CoM}$.

Once we have determined the center-of-mass-corrected renormalized Hamiltonian H, Eq. (4), we solve the nuclear many-body problem using coupled-cluster theory. To construct the coupled-cluster equations for the closed-shell A-body system and the related PA-EOMCC and PR-EOMCC equations

for the $(A+1)$- and $(A-1)$-body nuclei in the computationally most efficient way, we sort the one- and two-body matrix elements of H_N according to the particle-hole (p-h) character of the single-particle indices that label them prior to the coupled-cluster work. This is a common practice in the most efficient implementations of coupled-cluster methods by quantum chemists and we follow the same recipe here.

B. Brief synopsis of the single-reference coupled-cluster theory and the basic CCSD approximation

The single-reference coupled-cluster theory [10–13] is based on the exponential ansatz for the ground-state wave function of the A-body system,

$$\left|\Psi_0^{(A)}\right\rangle = e^{T^{(A)}}|\Phi\rangle, \tag{7}$$

where $T^{(A)}$ is the cluster operator (a p-h excitation operator) and $|\Phi\rangle$ is the corresponding reference determinant (defining the Fermi vacuum) obtained by performing some mean-field calculation or by simply filling A lowest-energy single-particle states (this is what we have done in the calculations discussed in this article). Here and elsewhere in the present article, we use superscripts, such as (A), which indicate the number of particles in a system under consideration, at the relevant operators and energies.

Formally, Eq. (7) is a direct consequence of the connected-cluster theorem, first clearly stated by Hubbard [63], which is, in turn, related to the linked cluster theorem of many-body perturbation theory [63–66]. According to the connected-cluster theorem, the cluster operator $T^{(A)}$ generates connected wave function diagrams summed through infinite order. Operationally, $T^{(A)}$ is a simple many-body excitation operator, which in all standard coupled-cluster approximations is truncated at a given (usually low) p-h excitation level $M<A$.

The general form of the truncated cluster operator, defining a standard single-reference coupled-cluster approximation characterized by the excitation level M, is

$$T^{(A)}(M) = \sum_{n=1}^{M} T_n, \tag{8}$$

where

$$T_n = \left(\frac{1}{n!}\right)^2 t^{i_1\ldots i_n}_{a_1\ldots a_n} a^{a_1}\ldots a^{a_n} a_{i_n}\ldots a_{i_1} \tag{9}$$

are the many-body components of $T^{(A)}(M)$ and $t^{i_1\ldots i_n}_{a_1\ldots a_n}$ are the corresponding cluster amplitudes. The cluster amplitudes $t^{i_1\ldots i_n}_{a_1\ldots a_n}$ are determined by solving a coupled system of nonlinear and energy-independent algebraic equations of the form:

$$\left\langle \Phi^{a_1\ldots a_n}_{i_1\ldots i_n}\right| \bar{H}_N(M)|\Phi\rangle = 0, \quad i_1<\cdots<i_n, \quad a_1<\cdots<a_n, \tag{10}$$

where $n=1,\ldots,M$. Here,

$$\bar{H}_N(M) = e^{-T^{(A)}(M)} H_N e^{T^{(A)}(M)} = \left(H_N e^{T^{(A)}(M)}\right)_C \tag{11}$$

is the similarity-transformed Hamiltonian of the coupled-cluster theory truncated at Mp-Mh excitations, subscript C designates the connected part of the corresponding operator expression, and $|\Phi^{a_1\ldots a_n}_{i_1\ldots i_n}\rangle \equiv a^{a_1}\ldots a^{a_n}a_{i_n}\ldots a_{i_1}|\Phi\rangle$ are the np-nh or n-tuply excited determinants relative to $|\Phi\rangle$. The basic CCSD (coupled-cluster singles and doubles) [67–69] method corresponds to $M=2$ and the cluster operator $T^{(A)}$ is approximated by

$$T^{(A)}(\mathrm{CCSD}) \equiv T^{(\mathrm{A})}(2) = T_1 + T_2, \tag{12}$$

where

$$T_1 = t^i_a a^a a_i \tag{13}$$

and

$$T_2 = \tfrac{1}{4} t^{ij}_{ab} a^a a^b a_j a_i \tag{14}$$

are $1p$-$1h$ or singly excited and $2p$-$2h$ or doubly excited cluster components, t^i_a and t^{ij}_{ab} are the corresponding singly and doubly excited cluster amplitudes, and $i, j, \ldots$ $(a, b, \ldots)$ are the single-particle states occupied (unoccupied) in the reference determinant $|\Phi\rangle$.

The standard CCSD equations for the singly and doubly excited cluster amplitudes t^i_a and t^{ij}_{ab}, defining T_1 and T_2, respectively, can be written as

$$\left\langle \Phi^a_i\right| \bar{H}_N(\mathrm{CCSD})|\Phi\rangle = 0, \tag{15}$$

$$\left\langle \Phi^{ab}_{ij}\right| \bar{H}_N(\mathrm{CCSD})|\Phi\rangle = 0, \qquad \mathrm{i<j}, \quad \mathrm{a<b}, \tag{16}$$

where

$$\begin{aligned}\bar{H}_N(\mathrm{CCSD}) \equiv \bar{H}_N(2) &= e^{-T^{(A)}(\mathrm{CCSD})} H_N e^{T^{(A)}(\mathrm{CCSD})} \\ &= \left(H_N e^{T^{(A)}(\mathrm{CCSD})}\right)_C\end{aligned} \tag{17}$$

is the similarity-transformed Hamiltonian of the CCSD approach.

The system of coupled-cluster equations, Eq. (10), is obtained in the following way (suggested by Čížek [12]). We first insert the coupled-cluster wave function $|\Psi_0^{(A)}\rangle$, Eq. (7), into the A-body Schrödinger equation,

$$H_N \left|\Psi_0^{(A)}\right\rangle = \Delta E_0^{(A)} \left|\Psi_0^{(A)}\right\rangle, \tag{18}$$

where

$$\Delta E_0^{(A)} = E_0^{(A)} - \langle\Phi|H|\Phi\rangle \tag{19}$$

is the corresponding energy relative to the reference energy $\langle\Phi|H|\Phi\rangle$, and premultiply both sides of Eq. (18) on the left by $e^{-T^{(A)}}$ to obtain the connected-cluster form of the Schrödinger equation [12,14,20,70],

$$\bar{H}_N|\Phi\rangle = \Delta E_0^{(A)}|\Phi\rangle\ , \tag{20}$$

where

$$\bar{H}_N = e^{-T^{(A)}} H_N e^{T^{(A)}} = \left(H_N e^{T^{(A)}}\right)_C \tag{21}$$

is the similarity-transformed Hamiltonian. Next, we project Eq. (20), in which $T^{(A)}$ is replaced by its approximate form $T^{(A)}(M)$, Eq. (8), onto the excited determinants $|\Phi^{a_1\ldots a_n}_{i_1\ldots i_n}\rangle$, corresponding to the p-h excitations included in $T^{(A)}(M)$. The excited determinants $|\Phi^{a_1\ldots a_n}_{i_1\ldots i_n}\rangle$ are orthogonal to the reference determinant $|\Phi\rangle$, so that we end up with nonlinear

and energy-independent algebraic equations of the form of Eq. (10).

Once the system of equations, Eq. (10), is solved for $T^{(A)}(M)$ or $t^{i_1\ldots i_n}_{a_1\ldots a_n}$ [or Eqs. (15) and (16) are solved for T_1 and T_2 or t^i_a and t^{ij}_{ab}], the ground-state coupled-cluster energy is calculated using the equation

$$\begin{aligned} E_0^{(A)}(M) &= \langle\Phi|H|\Phi\rangle + \Delta E_0^{(A)}(M) \\ &= \langle\Phi|H|\Phi\rangle + \langle\Phi|\bar{H}_N(M)|\Phi\rangle \\ &= \langle\Phi|H|\Phi\rangle + \langle\Phi|\bar{H}_{N,\mathrm{close}}(M)|\Phi\rangle, \end{aligned} \tag{22}$$

where $\bar{H}_{N,\mathrm{close}}(M)$ is the closed part of $\bar{H}_N(M)$ which is represented by those diagrams contributing to $\bar{H}_N^{(M)}$ that have no external (uncontracted) Fermion lines. It can easily be shown that if H contains only up to two-body interactions and $2 \leqslant M \leqslant A$, we can write

$$E_0^{(A)}(M) = \langle\Phi|H|\Phi\rangle + \langle\Phi|\left[H_N\left(T_1 + T_2 + \tfrac{1}{2}T_1^2\right)\right]_C|\Phi\rangle. \tag{23}$$

In other words, we need only T_1 and T_2 clusters to calculate the ground-state energy $E_0^{(A)}(M)$ of the A-body ($A \geqslant 2$) system even if we solve for other cluster components T_n with $n > 2$. As long as the Hamiltonian contains up to two-body interactions, the above energy expression is correct even in the exact case, when the cluster operator T is not truncated. In that case, however, to obtain the exact values of the T_1 and T_2 clusters and, thus, the exact ground-state energy, we would also need to solve the coupled system of equations for all higher-order clusters T_n with $n > 2$. Equation (22) can be most easily obtained by projecting the connected-cluster form of the Schrödinger equation, Eq. (20), on the reference configuration $|\Phi\rangle$ and replacing $T^{(A)}$ by $T^{(A)}(M)$.

The nonlinear character of the system of coupled-cluster equations of the form of Eq. (10) does not mean that the resulting equations contain very high powers of $T^{(A)}(M)$. For example, if the Hamiltonian H does not contain higher-than-pairwise interactions, the CCSD equations for the T_1 and T_2 clusters, or for the amplitudes t^i_a and t^{ij}_{ab} that represent these clusters, become

$$\left\langle\Phi_i^a\right|\left[H_N\left(1 + T_1 + T_2 + \tfrac{1}{2}T_1^2 + T_1T_2 + \tfrac{1}{6}T_1^3\right)\right]_C|\Phi\rangle = 0, \tag{24}$$

$$\begin{aligned} \left\langle\Phi_{ij}^{ab}\right|\Big[H_N\big(1 + T_1 + T_2 + \tfrac{1}{2}T_1^2 + T_1T_2 + \tfrac{1}{6}T_1^3 \\ + \tfrac{1}{2}T_2^2 + \tfrac{1}{2}T_1^2T_2 + \tfrac{1}{24}T_1^4\big)\Big]_C|\Phi\rangle = 0. \end{aligned} \tag{25}$$

The explicitly connected form of the coupled-cluster equations, such as Eqs. (10) or (24) and (25), guarantees that the process of solving these equations leads to connected terms in cluster components T_n and connected terms in the energy $E_0^{(A)}(M)$, independent of the truncation scheme M used to define $T^{(A)}(M)$. The absence of disconnected terms in $T^{(A)}(M)$ and $E_0^{(A)}(M)$ is essential to obtain the rigorously size-extensive results.

C. Equation-of-motion coupled-cluster methods for valence systems: the PA-EOMCCSD and PR-EOMCCSD approximations

In addition to providing natural intermediates for setting up coupled-cluster equations (see the Appendix), the use of the similarity-transformed Hamiltonians, $\bar{H}_N(M)$ or $\bar{H}_N(\mathrm{CCSD})$, Eqs. (11) or (17), respectively, in coupled-cluster calculations provides a natural mechanism for extending the ground-state coupled-cluster theory to excited states of a given A-body system or to ground and excited states of the $(A + k)$- or $(A - k)$-particle systems obtained by attaching k particles to or removing k particles from the A-particle closed-shell core. This can be most efficiently done by exploiting the EOMCC formalism [46,57,58] and its PA-EOMCC (in chemistry, EA-EOMCC [46–50]) and PR-EOMCC (in chemistry, IP-EOMCC [16,46,50–56]) variants, and their various multiply attached and multiply removed or ionized (cf., e.g., Refs. [50,71–73]) extensions (see, also, Refs. [74–78] and [79–84] for the related linear response and symmetry-adapted cluster configuration interaction formalisms, respectively). In all of these methods, we obtain excited states $|\Psi_\mu^{(A)}\rangle$ ($\mu > 0$) of the A-particle system or ground and excited states $|\Psi_\mu^{(A\pm k)}\rangle$ ($\mu \geqslant 0$) of the $(A \pm k)$-particle ($k > 0$) systems by applying the suitably defined excitation ($R_\mu^{(A)}$) or particle-attaching/particle-removing ($R_\mu^{(A\pm k)}$) operator to the ground state obtained in the single-reference coupled-cluster calculations for the closed-shell A-body system. Operators $R_\mu^{(A)}$ and $R_\mu^{(A\pm k)}$ are obtained by diagonalizing the similarity-transformed Hamiltonians, such as $\bar{H}_N(M)$, Eq. (11), in the case of coupled-cluster theory truncated at M-tuple excitations, or $\bar{H}_N(\mathrm{CCSD})$, Eq. (17), in the CCSD case, in the relevant A-particle and $(A \pm k)$-particle subspaces of the Fock space.

In the basic EOMCC approximation, EOMCCSD [57,58], for the calculations of excited states of the A-particle system, we represent excited states $|\Psi_\mu^{(A)}\rangle$ as

$$\left|\Psi_\mu^{(A)}\right\rangle = R_\mu^{(A)}\left|\Psi_0^{(A)}\right\rangle = R_\mu^{(A)}e^{T^{(A)}}|\Phi\rangle \tag{26}$$

and replace $T^{(A)}$ by the cluster operator $T^{(A)}(\mathrm{CCSD})$, Eq. (12), obtained in the CCSD calculations, and $R_\mu^{(A)}$ by

$$R_\mu^{(A)}(\mathrm{CCSD}) \equiv R_\mu^{(A)}(2) = R_{\mu,0} + R_{\mu,1} + R_{\mu,2}, \tag{27}$$

where

$$R_{\mu,0} = r_0\,1, \tag{28}$$

$$R_{\mu,1} = r_a^i\ a^a a_i, \tag{29}$$

and

$$R_{\mu,2} = \tfrac{1}{4}r_{ab}^{ij}\,a^a a^b a_j a_i \tag{30}$$

are the reference, $1p$-$1h$, and $2p$-$2h$ components of $R_\mu^{(A)}(\mathrm{CCSD})$, and r_0, r_a^i, and r_{ab}^{ij} are the corresponding excitation amplitudes [1 in Eq. (28) is a unit operator].

The r_a^i and r_{ab}^{ij} amplitudes of the standard EOMCCSD theory and the corresponding excitation energies $\omega_\mu^{(A)}(\mathrm{CCSD})$ of the A-body system are obtained by diagonalizing the open part of the similarity-transformed Hamiltonian of the CCSD

approach,

$$\begin{aligned}\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) &= \left(H_N e^{T^{(A)}(\mathrm{CCSD})}\right)_{C,\mathrm{open}} \\ &= e^{-T^{(A)}(\mathrm{CCSD})} H_N e^{T^{(A)}(\mathrm{CCSD})} - \bar{H}_{N,\mathrm{close}}(\mathrm{CCSD}) \\ &= e^{-T^{(A)}(\mathrm{CCSD})} H_N e^{T^{(A)}(\mathrm{CCSD})} - \Delta E_0^{(A)}(\mathrm{CCSD}), \end{aligned} \tag{31}$$

in the subspace spanned by the singly and doubly excited determinants $|\Phi_i^a\rangle$ and $|\Phi_{ij}^{ab}\rangle$ used to set up and solve the ground-state CCSD equations. The EOMCCSD approach can easily be generalized to higher-order excitation amplitudes by considering higher-than-two-body terms in $R_\mu^{(A)}$.

The idea of diagonalizing the similarity-transformed Hamiltonian of the coupled-cluster theory defining the EOM-CCSD and other EOMCC approximations can be extended to ground and excited states of open-shell nuclei with $(A \pm k)$ particles by replacing the particle-conserving p-h excitation operator $R_\mu^{(A)}$ in Eq. (26) by the suitably defined particle-attaching or particle-removing operator $R_\mu^{(A\pm k)}$. In the basic PA-EOMCCSD [47,48] and PR-EOMCCSD [16,51–53] approaches (cf., also, Ref. [46,50]) exploited in this work, we define the wave functions of the $(A+1)$- and $(A-1)$-particle systems, respectively, as

$$\left|\Psi_\mu^{(A\pm1)}\right\rangle = R_\mu^{(A\pm1)} e^{T^{(A)}} |\Phi\rangle, \tag{32}$$

where $T^{(A)}$ is approximated by $T^{(A)}(\mathrm{CCSD})$, Eq. (12), obtained in the CCSD calculations for the A-particle closed-shell system, and $R_\mu^{(A+1)}$ and $R_\mu^{(A-1)}$ are replaced by the appropriately truncated operators,

$$R_\mu^{(A+1)}(2p\text{-}1h) = R_{\mu,1p} + R_{\mu,2p\text{-}1h} = r_a a^a + \tfrac{1}{2} r_{ab}^j a^a a^b a_j \tag{33}$$

and

$$R_\mu^{(A-1)}(2h\text{-}1p) = R_{\mu,1h} + R_{\mu,2h\text{-}1p} = r^i a_i + \tfrac{1}{2} r_b^{ij} a^b a_j a_i, \tag{34}$$

respectively, which generate the $(A+1)$- and $(A-1)$-particle states from the A-particle CCSD wave function $e^{T_1+T_2}|\Phi\rangle$. The $1p$ and $2p$-$1h$ amplitudes r_a and r_{ab}^j, respectively, entering Eq. (33) and defining the PA-EOMCCSD model, and the $1h$ and $2h$-$1p$ amplitudes r^i and r_b^{ij}, respectively, entering Eq. (34) and defining the PR-EOMCCSD model, are determined by solving the eigenvalue problem,

$$\left(\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) R_\mu^{(A\pm1)}\right)_C |\Phi\rangle = \omega_\mu^{(A\pm1)} R_\mu^{(A\pm1)} |\Phi\rangle, \tag{35}$$

in the relevant subspaces of the $(A+1)$- and $(A-1)$-particle subspaces, $\mathcal{H}^{(A+1)}$ and $\mathcal{H}^{(A-1)}$, respectively, of the Fock space. The subspace of $\mathcal{H}^{(A+1)}$ used to solve the PA-EOMCCSD eigenvalue problem is spanned by the $|\Phi^a\rangle = a^a|\Phi\rangle$ and $|\Phi_j^{ab}\rangle = a^a a^b a_j |\Phi\rangle$ determinants. The subspace of $\mathcal{H}^{(A-1)}$ used to solve the PR-EOMCCSD problem is spanned by the $|\Phi_i\rangle = a_i|\Phi\rangle$ and $|\Phi_{ij}^b\rangle = a^b a_j a_i |\Phi\rangle$ determinants. By solving Eq. (35), we directly obtain the energy differences, $\omega_\mu^{(A+1)} = E_\mu^{(A+1)} - E_0^{(A)}$ in the PA-EOMCCSD case, and $\omega_\mu^{(A-1)} = E_\mu^{(A-1)} - E_0^{(A)}$ in the PR-EOMCCSD case, where $E_\mu^{(A+1)}$ and $E_\mu^{(A-1)}$ are the energies of ground ($\mu = 0$) and excited ($\mu > 0$) states of the $(A+1)$- and $(A-1)$-particle systems, respectively, and $E_0^{(A)}$ is the ground-state coupled-cluster (in this case, CCSD) energy of the A-particle reference system. Note that these methods naturally extend to higher-order excitations. The detailed discussion of the relationships between truncation schemes in the $R_\mu^{(A\pm1)}$ and $T^{(A)}$ operators in the PA-EOMCC and PR-EOMCC calculations can be found in Ref. [46] (cf., also, Refs. [50,85] for additional comments and numerical tests).

The PA-EOMCC and PR-EOMCC methods, as described above, and their extensions to two or more valence particle or holes via multiply attached or multiply ionized schemes [50,71–73] offer several advantages compared to the equally accurate, but usually much more complicated, genuine multireference coupled-cluster methods of either the valence-universal [86,87] or the Hilbert-space or state-universal [88] type that are specifically designed to handle general classes of open-shell problems. Although there has been significant progress in recent years in the development of genuine multireference coupled-cluster theories [89–101], multireference coupled-cluster calculations are often plagued by intruder states; unphysical, singular, and multiple solutions; and mathematical difficulties with the proper adaptation of the corresponding equations to symmetries of the Hamiltonian if one aims at the general-purpose computer codes (cf., e.g., Refs. [90,102–109] for further information). Some of these issues are currently being addressed (cf., e.g., Refs. [92–99,101]), but none of these problems are present in the PA-EOMCC and PR-EOMCC calculations, which could be viewed as the physically motivated, intruder-state-free, state-selective modifications of the powerful and elegant valence-universal multireference coupled-cluster schemes pioneered by Mukherjee and Lindgren [86,87].

Our calculations for the ground and low-lying excited states of the 15- and 17-particle nuclei around ^{16}O, reported in this work, have been performed with the basic PA-EOMCCSD and PR-EOMCCSD methods, in which the ground state of ^{16}O is represented by the CCSD wave function $e^{T_1+T_2}|\Phi\rangle$ and the nucleon-attaching and nucleon-removing operators $R_\mu^{(A+1)}$ and $R_\mu^{(A-1)}$ are defined by Eqs. (33) and (34), respectively. As a first approximation we can describe the $(A+1)$-particle nuclei ^{17}O and ^{17}F with the PA-EOMCCSD method, in which we include the $1p$ and $2p$-$1h$ excitations from the ^{16}O core to form the 17-particle systems, because the ground states of the ^{17}O and ^{17}F nuclei that we have singled out in this work are essentially one-quasiparticle states. On the other hand, the $(3/2)_1^+$ excited states of ^{17}O and ^{17}F are resonances that can strongly couple to more complicated excitations that are neglected in the PA-EOMCCSD calculations. This may apply to the $(1/2)_1^+$ states as well. We can, however, study the $(A-1)$-particle nuclei ^{15}O and ^{15}N with the basic PR-EOMCCSD approach, in which we include the $1h$ and $2h$-$1p$ excitations from the ^{16}O closed-shell core, because the low-lying states of these nuclei are expected to be dominated by one-quasihole states with respect to the A-body reference ^{16}O nucleus. As discussed in Ref. [1], there is, for example, almost no experimental evidence for the fragmentation of the quasihole $p_{1/2}$ and $p_{3/2}$ states of ^{16}O.

The fact that we use the $1p$ and $2p$-$1h$ excitations in the PA-EOMCCSD calculations to form the $(A+1)$-body systems and the fact that we use the $1h$ and $2h$-$1p$ excitations in the PR-EOMCCSD calculations for the $(A-1)$-body systems mean that we include many of the same correlations as Fujii *et al.* [110,111]. Their approach is analogous to a Hermitian coupled-cluster approach (see Ref. [111]). Another approach is the Green's function approach, see the work of Barbieri and Dickhoff [112]. There are, however, differences between our PA-EOMCCSD and PR-EOMCCSD calculations and the calculations reported by Fujii *et al.* In particular, we use a biorthogonal EOMCC formalism, based on diagonalizing the non-Hermitian similarity-transformed Hamiltonian $\bar{H}_N$(CCSD), Eq. (17), obtained in CCSD calculations for the A-body closed-shell nucleus, which brings a lot of correlations within basic truncation schemes, such as EOMCCSD, PA-EOMCCSD, and PR-EOMCCSD, through the presence of high-order correlation terms in $\bar{H}_N$(CCSD).

We also differ in the definition of the model space, since Fujii *et al.* use a model space similar to that used in the no-core shell-model calculations [4], in which a "triangular" energy cutoff is applied to Slater determinants included in the diagonalization of the Hamiltonian, in addition to the usual single-particle basis set cutoff. Such a model space cannot be used in coupled-cluster calculations because it violates the Pauli principle in the summations over the intermediate states that emerge through products of many-body components of the cluster operator $T^{(A)}$ in coupled-cluster equations. As mentioned earlier, the use of a given truncation scheme for the cluster operator $T^{(A)}$ implies specific truncation schemes for the EOMCC operators, such as $R_\mu^{(A+1)}$ and $R_\mu^{(A-1)}$. Thus, we use all $1p$ and $2p$-$1h$ or $1h$ and $2h$-$1p$ excitations in the PA-EOMCCSD and PR-EOMCCSD calculations and all $1p$-$1h$ and $2p$-$2h$ cluster amplitudes t_a^i and t_{ab}^{ij} that are allowed by a given single-particle basis set, without imposing additional energy cutoffs on the determinants that these excitations correspond to, producing many additional and important correlations that are outside model spaces used in the no-core shell-model calculations. In principle, however, our approach will miss center-of-mass excitations that will be included in a translationally invariant no-core shell-model calculation, if all excitations are allowed in large spaces. As we show below, however, the expectation value of spurious center-of-mass components become negligible as we increase the size of the model space.

D. Further computational details of the PA-EOMCCSD and PR-EOMCCSD calculations

Once the one- and two-body matrix elements of the center-of-mass-corrected effective Hamiltonian, Eq. (4), are determined and properly sorted out, we set up and solve the CCSD equations for ^{16}O, the PA-EOMCCSD equations for ^{17}O and ^{17}F, and the PR-EOMCCSD equations for ^{15}O and ^{15}N. Our ground-state CCSD computer codes rely on the DIIS solver [113] (see, also, Refs. [44,114]), whereas the PA-EOMCCSD and PR-EOMCCSD equations for ground and excited states of the $(A+1)$- and $(A-1)$-particle nuclei are solved with the Hirao-Nakatsuji generalization [115] of the Davidson diagonalization algorithm [116] to non-Hermitian eigenvalue problems.

The computationally efficient form of the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations in terms of recursively generated intermediates can be derived diagrammatically. From the point of view of code efficiency, it is important to realize that some of the intermediates entering the CCSD and other CCSD-based equations represent matrix elements of the one- and two-body components of the CCSD similarity-transformed Hamiltonian $\bar{H}_{N,\mathrm{open}}$(CCSD), Eq. (31). If $\bar{H}_n$ is the n-body component of $\bar{H}_{N,\mathrm{open}}$(CCSD), for the one- and two-body components $\bar{H}_1$ and $\bar{H}_2$, respectively, we can write

$$\bar{H}_1 = \bar{h}_\alpha^\beta N[a^\alpha a_\beta] \tag{36}$$

and

$$\bar{H}_2 = \tfrac{1}{4}\bar{h}_{\alpha\beta}^{\gamma\delta} N[a^\alpha a^\beta a_\delta a_\gamma], \tag{37}$$

where $\bar{h}_\alpha^\beta$ and $\bar{h}_{\alpha\beta}^{\gamma\delta}$ are the one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}$(CCSD) that enter the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations. As shown in the Appendix, matrix elements $\bar{h}_\alpha^\beta$ and $\bar{h}_{\alpha\beta}^{\gamma\delta}$ are calculated using the one- and two-body matrix elements of the Hamiltonian in the normal-ordered form, f_α^β and $v_{\alpha\beta}^{\gamma\delta}$, respectively [cf. Eq. (5)], and the singly and doubly excited cluster amplitudes t_a^i and t_{ab}^{ij}, defining T_1 and T_2, respectively. The computationally efficient form of the CCSD equations for the case of pairwise interactions in H, in terms of selected types of $\bar{h}_\alpha^\beta$ and $\bar{h}_{\alpha\beta}^{\gamma\delta}$ and other recursively generated intermediates, is given in the Appendix.

We also give in the Appendix the computationally efficient form of the equations defining the PA-EOMCCSD and PR-EOMCCSD eigenvalue problems. These are obtained by applying diagrammatic methods to the PA-EOMCCSD and PR-EOMCCSD equations, which can be given the following general form:

$$\langle\Phi^a|\left[(\bar{H}_1 R_{\mu,1p})_C + \sum_{n=1}^{2}(\bar{H}_n R_{\mu,2p\text{-}1h})_C\right]|\Phi\rangle = \omega_\mu^{(A+1)} r_a, \tag{38}$$

$$\left\langle\Phi_j^{ab}\right|\left[(\bar{H}_2 R_{\mu,1p})_C + \sum_{n=1}^{3}(\bar{H}_n R_{\mu,2p\text{-}1h})_C\right]|\Phi\rangle = \omega_\mu^{(A+1)} r_{ab}^j, \tag{39}$$

in the PA-EOMCCSD case, and

$$\langle\Phi_i|\left[(\bar{H}_1 R_{\mu,1h})_C + \sum_{n=1}^{2}(\bar{H}_n R_{\mu,2h\text{-}1p})_C\right]|\Phi\rangle = \omega_\mu^{(A-1)} r^i, \tag{40}$$

$$\left\langle\Phi_{ij}^{b}\right|\left[(\bar{H}_2 R_{\mu,1h})_C + \sum_{n=1}^{3}(\bar{H}_n R_{\mu,2h\text{-}1p})_C\right]|\Phi\rangle = \omega_\mu^{(A-1)} r_b^{ij}, \tag{41}$$

in the PR-EOMCCSD case. Although formally the PA-EOMCCSD and PR-EOMCCSD equations require the consideration of the three-body components of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$ [cf. the $n = 3$ terms in Eqs. (39) and (41)], we do not have to calculate the corresponding six-index matrix elements $\bar{h}^{\delta\epsilon\eta}_{\alpha\beta\gamma}$ explicitly. With the help of diagrammatic techniques, the three-body components of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$ that enter the PA-EOMCCSD and PR-EOMCCSD equations can be rigorously factorized and rewritten in terms of the one- and two-body components of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$. In consequence, the final working equations of the PA-EOMCCSD and PR-EOMCCSD methods in terms of one- and two-body matrix elements of the Hamiltonian, f^{β}_{α} and $v^{\gamma\delta}_{\alpha\beta}$, respectively, T_1 and T_2 cluster amplitudes defining the underlying A-particle ground-state CCSD problem, and the $R_{\mu,1p}$, $R_{\mu,2p\text{-}1h}$, $R_{\mu,1h}$, and $R_{\mu,2h\text{-}1p}$ excitation amplitudes defining the particle-attaching and particle-removing operators, $R^{(A+1)}_{\mu}(2p\text{-}1h)$ and $R^{(A-1)}_{\mu}(2h\text{-}1p)$, respectively, can be reexpressed in terms of the one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$, $\bar{h}^{\beta}_{\alpha}$ and $\bar{h}^{\gamma\delta}_{\alpha\beta}$, respectively, and a few additional recursively generated intermediates, leading to a fully vectorizable algorithm.

In addition to code vectorization, another advantage of deriving the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations in the form shown in the Appendix is the possibility of obtaining the relatively low CPU operation count that characterizes these methods. The CCSD equations and the determination of the full set of one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$ are characterized by the $n_o^2 n_u^4$ steps, where n_o and n_u are the numbers of occupied and unoccupied orbitals, respectively, in the single-particle basis set. Once the CCSD equations are solved and all one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$ are determined, the most expensive steps of the PA-EOMCCSD and PR-EOMCCSD methods employing the factorized equations shown in the Appendix are $n_o n_u^4$ and $n_o^2 n_u^3$, respectively. These relatively low, $\mathcal{N}^5 - \mathcal{N}^6$ scalings of the costs of the CCSD, PA-EOMCCSD, and PR-EOMCCSD calculations with the system size ($\mathcal{N}$), which are often orders of magnitude smaller than the costs of shell-model calculations aimed at similar accuracies, are among the most important advantages of the coupled-cluster methodology pursued in this work.

III. RESULTS AND DISCUSSION

A. Results of the PR-EOMCCSD and PA-EOMCCSD calculations with the N^{3}LO interaction and their convergence properties

We focus first on the convergence of our PR-EOMCCSD and PA-EOMCCSD results for the ground and excited states of ^{15}O, ^{15}N, ^{17}O, and ^{17}F with the size of the single-particle basis set used in the coupled-cluster calculations and address the issue of the dependence of these results on the choice of the oscillator parameter $\hbar\Omega$ (see Tables I – V). For comparison purposes, we also list our previously published ground-state CCSD results with $\langle T_{\mathrm{CoM}}\rangle$ subtracted for ^{16}O [38], because ^{16}O serves as a reference nucleus for the PR-EOMCCSD and PA-EOMCCSD calculations. We limit our discussion of the convergence properties of the PR-EOMCCSD and PA-EOMCCSD results for ^{15}O, ^{15}N, ^{17}O, and ^{17}F to the N^{3}LO interaction model [31]. The PR-EOMCCSD and PA-EOMCCSD results for the CD-Bonn [29] and Argonne V_{18} [28] interactions exhibit almost identical qualitative features in terms of their convergence with the number of major oscillator shells and the way they depend on $\hbar\Omega$.

TABLE I. Total binding energies and binding energies per particle (in parentheses) for ^{15}O and ^{15}N (the PR-EOMCCSD values), ^{16}O (the CCSD values), and ^{17}O and ^{17}F (the PA-EOMCCSD values), computed with the N^{3}LO interaction model [31], as functions of the number of major oscillator shells N. All entries (except for the unitless parameter β_{CoM}) are in MeV. The results for ^{16}O are taken from Ref. [38]. The acronym Expt. stands for the experimental values, taken from Ref. [117]. All energies were calculated at the optimum values of $\hbar\Omega$ (the second last row; determined by identifying the $\hbar\Omega$ value at which the CCSD energy of ^{16}O reaches the minimum value) and β_{CoM} (the last row; determined by the condition that the expectation value of H_{CoM} with the CCSD wave function is 0.0 MeV). For eight major oscillator shells, $\beta_{\mathrm{CoM}} = 0.0$.

Nucleus	$N = 5$	$N = 6$	$N = 7$	$N = 8$	Expt.
^{15}O	113.904	101.151	99.869	99.646	111.955
	(7.594)	(6.743)	(6.658)	(6.643)	(7.464)
^{15}N	116.983	103.811	102.335	102.360	115.492
	(7.799)	(6.921)	(6.823)	(6.824)	(7.699)
^{16}O	131.800	120.720	119.096	118.491	127.619
	(8.787)	(7.545)	(7.444)	(7.406)	(7.976)
^{17}O	132.075	122.913	121.977	121.547	131.762
	(8.805)	(7.230)	(7.175)	(7.150)	(7.751)
^{17}F	128.954	120.248	119.432	118.780	128.220
	(8.597)	(7.073)	(7.026)	(6.987)	(7.542)
$\hbar\Omega$	13	11	10	11	
β_{CoM}	1.50	0.15	0.05	0.0	

As shown in Table I, the PR-EOMCCSD binding energies of ^{15}O and ^{15}N, the results of the CCSD calculations for the binding energy of ^{16}O, and the PA-EOMCCSD binding energies of ^{17}O and ^{17}F are practically converged at the level of eight major oscillator shells. As demonstrated earlier for

TABLE II. Dependence of the total binding energies on $\hbar\Omega$ for ^{15}O and ^{15}N (the PR-EOMCCSD values), ^{16}O (the CCSD values), and ^{17}O and ^{17}F (the PA-EOMCCSD values), computed with the N^{3}LO interaction model [31] for $N = 6$ and $N = 7$ oscillator shells. All entries (except for the unitless parameter β_{CoM}) are in MeV. The optimum value of β_{CoM} is given in the last row.

Nucleus	$N = 6$	$N = 6$	$N = 6$	$N = 7$	$N = 7$
^{15}O	101.151	100.731	99.708	99.869	95.328
^{15}N	103.811	103.731	103.034	102.341	98.686
^{16}O	120.720	119.781	118.171	119.096	113.864
^{17}O	122.913	121.952	120.082	121.977	116.409
^{17}F	120.248	118.977	116.792	119.432	113.230
$\hbar\Omega$	11	12.5	14	10	14
β_{CoM}	0.15	0.3	0.5	0.05	0.3

TABLE III. Energies of the low-lying excited states of ^{15}O, ^{15}N, ^{17}O, and ^{17}F, relative to the corresponding ground-state energies [the $(1/2)^-_1$ states of ^{15}O and ^{15}N and the $(5/2)^+_1$ states of ^{17}O and ^{17}F], computed with the N^{3}LO interaction model [31] and the PR-EOMCCSD (^{15}O,^{15}N) and PA-EOMCCSD (^{17}O and ^{17}F) methods, as functions of the number of major oscillator shells N. All entries are in MeV. Note that the experimentally observed $(3/2)^+_1$ states in ^{17}O and ^{17}F are resonances. The experimental data (Expt) are from Ref. [121]. For the optimum values of $\hbar\Omega$ and β_{CoM}, see Table I.

Excited state	$N=5$	$N=6$	$N=7$	$N=8$	Expt.
^{15}O $(3/2)^-_1$	6.515	6.602	6.166	6.264	6.176
^{15}N $(3/2)^-_1$	6.354	6.680	6.256	6.318	6.323
^{17}O $(3/2)^+_1$	6.298	6.031	5.489	5.675	5.084
^{17}O $(1/2)^+_1$	0.328	0.130	−0.349	−0.025	0.870
^{17}F $(3/2)^+_1$	6.460	6.207	5.686	5.891	5.000
^{17}F $(1/2)^+_1$	0.748	0.544	0.088	0.428	0.495

^{16}O [36], the dependence of these results on the oscillator energy $\hbar\Omega \in [10, 20]$ MeV is small, particularly for seven or eight major oscillator shells. However, because the Coulomb interaction is included perturbatively in all calculations, this leads to a weak dependence on the oscillator energy when we compare the binding energy difference between ^{15}O and ^{15}N and ^{17}O and ^{17}F. The not renormalized two-body Coulomb contribution increases with increasing $\hbar\Omega$ (to see this it suffices to set up the Coulomb integral in a harmonic oscillator basis). The reader should keep in mind that for every given oscillator energy our results are converged at the level of $N = 8$ major shells.

In Table II we display the dependence of the ground-state energy on $\hbar\Omega$ for $N = 6$ and $N = 7$ major shells for the nuclei investigated here. Note that the $A = 15$ nuclei show less energy dependence as a function of $\hbar\Omega$ than their $A = 17$ counterparts. We see from this Table that for six major shells and $\hbar\Omega = 14$ MeV we have a binding energy difference BE(^{15}N)−BE(^{15}O) = 3.326 MeV and BE(^{17}O)−BE(^{17}F) = 3.290 MeV, to be compared with the experimental values BE(^{15}N)−BE(^{15}O) = 3.537 MeV and BE(^{17}O)−BE(^{17}F) = 3.542 MeV. We note that the BE(^{15}N) − BE(^{15}O) = 2.47 and 2.71 MeV in the $N = 7, \hbar\Omega = 10$ MeV, and $N = 8, \hbar\Omega = 11$ MeV calculations, respectively. (Similar results hold for the ^{17}O, ^{17}F binding energy difference.) This

TABLE IV. Dependence on $\hbar\Omega$ of the excited state spectrum for the $N = 6$ model space, computed with N^{3}LO at the optimal values of β_{CoM}, which are $\beta_{\mathrm{CoM}} = 0.05, 0.3$, and 0.5, for $\hbar\Omega = 10$, 12.5, and 14 MeV, respectively.

Excited state	$\hbar\Omega = 11$	$\hbar\Omega = 12.5$	$\hbar\Omega = 14$	Expt.
^{15}O $(3/2)^-_1$	6.602	6.841	6.990	6.176
^{15}N $(3/2)^-_1$	6.680	6.889	7.034	6.323
^{17}O $(3/2)^+_1$	6.031	6.536	7.129	5.084
^{17}O $(1/2)^+_1$	0.130	0.440	1.219	0.870
^{17}F $(3/2)^+_1$	6.207	6.740	7.199	5.000
^{17}F $(1/2)^+_1$	0.544	0.884	1.219	0.495

TABLE V. Dependence on $\hbar\Omega$ of the excited state spectrum for the $N = 7$ model space, computed with N^{3}LO at the optimal values of β_{CoM}, which are $\beta_{\mathrm{CoM}} = 0.05$ for $\hbar\Omega = 11$ MeV and $\beta_{\mathrm{CoM}} = 0.3$ for $\hbar\Omega = 14$ MeV.

Excited state	$\hbar\Omega = 11$	$\hbar\Omega = 14$	Expt.
^{15}O $(3/2)^-_1$	6.166	6.668	6.176
^{15}N $(3/2)^-_1$	6.256	6.703	6.323
^{17}O $(3/2)^+_1$	5.489	6.419	5.084
^{17}O $(1/2)^+_1$	−0.349	0.481	0.870
^{17}F $(3/2)^+_1$	5.686	6.672	5.000
^{17}F $(1/2)^+_1$	0.088	1.002	0.495

observable clearly shows that either we need to treat the Coulomb as more than a perturbative quantity or that we still have some way to go to obtain a total convergence with oscillator space. The Coulomb energy difference is actually a derivative quantity and so will be more difficult to converge than the energies themselves. The charge radius for the ground state of ^{16}O, which is our reference system, also does not change much. The charge radius for six major shells changes from 2.647 to 2.572 fm for $\hbar\Omega = 11$ and 14 MeV, respectively. For seven major shells, the result for $\hbar\Omega = 14$ MeV is 2.595 fm, indicating a good convergence of the underlying CCSD calculations for ^{16}O.

Except for our perturbative treatment of the Coulomb interaction, our results indicate that the renormalization of the short-range part of the nucleon-nucleon interaction with the no-core G-matrix approach combined with the inclusion of singly and doubly excited clusters and the corresponding $1p$, $2p$-$1h$, $1h$, and $2h$-$1p$ excitations in the coupled-cluster and PR-EOMCC/PA-EOMCC calculations for the valence systems around ^{16}O leads to reasonably well converged ground-state energies of these systems. It is true that the N^{3}LO interaction model has a rather soft core, since it carries a cutoff in relative momentum of $\Lambda = 500$ MeV. Thus, in developing the effective two-body interaction based on N^{3}LO by diagonalizing the deuteron in an oscillator basis, one obtains a converged result to six leading digits with 50 to 60 oscillator shells for $\hbar\Omega \in [5, 50]$ MeV. For the CD-Bonn and V_{18} interactions, one needs more than 100 major shells to obtain a converged result for the deuteron. However, the advantage of the G-matrix approach used in this work is that we can renormalize the short-range part of the interaction exactly, because the free part of the G matrix is computed in a momentum basis first, with the relative momenta $|\mathbf{p}| \in [0, \infty)$. Thus, the renormalization problems of the short-range part of the two-body interaction, seen, for example, in the no-core approach [4], with a relatively slow convergence as a function of the harmonic oscillator excitations, are not present here. This means, in turn, that when we use this G matrix in coupled-cluster calculations, the results for all modern nucleon-nucleon potentials, such as N^{3}LO, CD-Bonn, and V_{18} used here, are converged for ^{16}O within eight major shells, although some slight $\hbar\Omega$ dependence may remain.

As shown in Table I, our coupled-cluster calculations miss the experimental binding energies (taken from Ref. [117]) by approximately 0.8–0.9 MeV per nucleon for the $A = 15$ nuclei

and by approximately 0.4–0.6 MeV per nucleon for the $A = 16$ and $A = 17$ systems. Several factors can contribute to these differences, but we believe that the three-body interactions are the primary source. It is true, for example, that we are using the solution to a two-body problem (our G matrix) as the starting point for defining a many-body Hamiltonian with pairwise interactions for the $A = 15$–17 nuclei, and it is known that a two-body interaction derived from the diagonalization of a three-body problem is different from the corresponding two-body interaction derived by diagonalizing the two-body problem (e.g., deuteron) [4,118,119]. However, as the size of the model space is increased, both two-body interactions yield very similar results (see the discussion in Ref. [118]). Because we use large model spaces with seven or even eight major oscillator shells, the differences between these two types of effective two-body interactions are minimal and cannot, as such, contribute to the differences between the coupled-cluster and experimental data observed in our calculations.

We are missing some correlations in our coupled-cluster calculations, which ignore, for example, T_3 clusters in the ground state calculations for ^{16}O and $3p$-$2h$ and $3h$-$2p$ components of $R_\mu^{(A+1)}$ and $R_\mu^{(A-1)}$ in the PR-EOMCC and PA-EOMCC calculations for the 15- and 17-particle nuclei. We earlier reported that for the ^{16}O ground state (see Refs. [38,40,41]) the T_3 clusters bring in at most a total of 1 MeV (less than 0.1 MeV per nucleon) and cannot, therefore, account for the observed differences between the binding energies per nucleon.

We also see indications that the ground states in the ^{15}O/^{15}N and ^{17}O/^{17}F systems are relatively stable as a function of increasing model space size, although there is still some $\hbar\Omega$ dependence, particularly in the $A = 17$ systems. Part of this dependence comes from our treatment of the Coulomb interaction, but the one-particle ground states may be more affected by higher-order correlations in which the $3p$-$2h$ terms play a larger role. Experimentally, the one-particle states in the $A = 17$ nuclei are more fragmented than the one-hole states found in the $A = 15$ systems.

We can thus summarize this part of our discussion by stating that much of the discrepancy between experiment and theory observed in Table I may be ascribed to the missing three-body interactions, which are not included in our effective Hamiltonians. One advantage of the nuclear interaction models based on effective field theory is that they allow for a consistent derivation of three-body terms (see, for example, Refs. [31,120]), and an exploration of such interactions in the coupled-cluster context would be very interesting.

We end this subsection by tabulating the results of the PR-EOMCCSD and PA-EOMCCSD calculations for the low-lying excited states of ^{15}O, ^{15}N, ^{17}O, and ^{17}F obtained with the N^{3}LO potential (see Table III; the experimental data are taken from Ref. [121]). Except for the $(3/2)_1^+$ resonance states in ^{17}O and ^{17}F, the other states listed in Table III are expected to be strongly dominated by one quasiparticle or quasihole states, meaning that the inclusion of the $1p$ and $2p$-$1h$ excitations in the PA-EOMCCSD calculations and the $1h$ and $2h$-$1p$ correlations in the PR-EOMCCSD calculations should provide a reasonable description of these states. This is confirmed in Table III. The PR-EOMCCSD/N^{3}LO results for the $(3/2)_1^-$ states of ^{15}O and ^{15}N, employing seven or eight major oscillator shells, are particularly impressive, producing errors relative to experiment that do not exceed 0.1 MeV. For the $(1/2)_1^+$ states we note, however, that our results are not fully converged. This could indicate that one may need more than $2p$-$1h$ excitations in the particle-attaching operator used in the PA-EOMCC calculations for these states as well.

In Tables IV and V we indicate the dependence of the excited states on the oscillator energy $\hbar\Omega$ for the $N = 6$ and $N = 7$ shell-model spaces, respectively. For $N = 6$, the excited states of the $A = 15$ system change by approximately 0.4 MeV in the $\hbar\Omega = 11$–14 MeV window, whereas the $A = 17$ states change by approximately 0.7–1.1 MeV in the same $\hbar\Omega$ window. This would indicate a stronger dependence on $\hbar\Omega$ for the $A = 17$ nuclei than for the states in the $A = 15$ and ^{16}O nuclei. We see a similar but decreasing dependence in the $N = 7$ calculations, where we only performed the check at $\hbar\Omega = 14$ MeV. The above tables demonstrate that at least for the hole states we obtain results that stabilize as function of the number of shells. For the excited states of ^{17}O and ^{17}F there is still a relatively strong dependence on the number of shells and $\hbar\Omega$. The $(3/2)_1^+$ states are known resonances and we do therefore expect that our approximation at the PA-EOMCCSD level may miss some important correlations in this case. This seems to apply to the $(1/2)_1^+$ states as well.

Within a single-particle picture, the splitting between the $(3/2)_1^-$ excited and $(1/2)_1^-$ ground states in ^{15}O and ^{15}N and the splitting between the $(3/2)_1^+$ excited and $(5/2)_1^+$ ground states in ^{17}O and ^{17}F should receive important contributions from the nuclear spin-orbit force. It is interesting to analyze to what extent the three-nucleon interactions may affect these splittings. At least for hole states there is strong evidence for this behavior, see Refs. [1,2]. The nucleon-nucleon interaction contains a short-range spin-orbit force, which in a meson-exchange model picture originates from heavier vector mesons. Several partial waves receive significant contributions from the two-body spin-orbit force. For example, the $^3P_2$ partial wave, crucial for the pairing properties in nuclei and neutron star matter, yields an attractive interaction up to almost 1 GeV in laboratory energy for the two-nucleon scattering. This attraction arises from the two-body spin-orbit force, because both the central and tensor force contributions are repulsive. Within the framework of many-body perturbation theory, the largest contribution to the spin-orbit force arises from the first-order Hartree-Fock diagram. Indeed, for the N^{3}LO model used here and for an oscillator energy $\hbar\Omega = 14$ MeV, we obtain an excitation energy of 5.412 MeV for the $0p_{3/2}$ state of ^{15}O, in reasonable agreement with the experimental and coupled-cluster data in Table III [122]. At the Hartree-Fock diagram level, the origin of the spin-orbit splitting comes then from the renormalization of the short range two-body spin-orbit force. The nuclear tensor force gives also, as a second- and higher-order process, a contribution to the single-particle spin-orbit splitting (see the detailed discussion in Ref. [2] for further information). The authors of Ref. [2] show how the second-order diagrams in many-body perturbation theory with the $2h$-$1p$ and $2p$-$1h$ intermediate states yield repulsive and attractive contributions to the single-

particle energies, respectively. Depending on the strength of the nuclear tensor force, the spin-orbit splittings can then be enhanced or reduced. If the tensor force is weak, as is the case for the N^{3}LO model, the reduced higher-order quenching of the tensor force terms enhances the spin-orbit splitting with respect to the Hartree-Fock diagram. Anticipating the discussion in Sec. III C, potentials with a stronger tensor force, such as the V_{18} model of the Argonne group [28], result in a smaller spin-orbit splitting than the N^{3}LO model [and a reduction in the $(3/2)^-_1 - (1/2)^-_1$ and $(3/2)^+_1 - (5/2)^+_1$ spacings in the ^{15}O/^{15}N and ^{17}O/^{17}F nuclei, respectively]. The authors of Refs. [1,2] demonstrated then that a two-pion three-nucleon interaction also contributes to the spin-orbit splitting. With the inclusion of such a term, Pieper and Pandharipande [1], reproduced very well the $(3/2)^-_1 - (1/2)^-_1$ splitting in ^{15}N. These findings were later corroborated by Heisenberg and Mihaila in their coupled-cluster calculations with three-body interactions for ^{16}O (see Refs. [32–35] and the discussion in the next subsection as well). The fact that we reproduce very well the experimental $(3/2)^-_1 - (1/2)^-_1$ spin-orbit splittings in ^{15}N and ^{15}O with the pairwise N^{3}LO model may indicate that the spin-orbit force associated with an eventual three-body force for N^{3}LO should be small. This may be an important finding for our understanding of the role of three-body forces in nuclear structure calculations. Whether this conclusion pertains to spectra of open-shell nuclei as well is worth further study. The no-core shell-model calculations of Ref. [4] for ^{6}Li and ^{10}Be with the N^{3}LO interaction with pertinent three-body interactions indicate larger three-body contributions.

In Sec. III C, we present results for the binding energies and spectra of the ^{15}N and ^{15}O nuclei and their ^{17}F and ^{17}O counterparts using the CD-Bonn [29] and the Argonne V_{18} [28] interaction models as well in order to see how much the effects due to three-body interactions may depend on the underlying two-body forces. However, before we proceed, let us discuss interesting consequences of our PR-EOMCCSD and PA-EOMCCSD calculations for ^{15}N, ^{15}O, ^{17}F, and ^{17}O for the nuclear structure studies of the excited states of ^{16}O.

B. Consequences of the PR-EOMCCSD and PA-EOMCCSD calculations for the valence systems around ^{16}O for the studies of excitations in ^{16}O

Based on the N^{3}LO results discussed in the previous subsection, we attempt to link our findings to nuclear structure studies of the excitations in ^{16}O. The fact that we obtain nearly converged results for a given two-body Hamiltonian allows us to infer that eventual disagreements with experiment in the results of *ab initio* calculations for excited states of ^{16}O can very likely be retraced to the degrees of freedom that are not included in the existing two-body Hamiltonians.

Here we discuss the excited states of ^{16}O with an expected $1p$-$1h$ structure. In our calculations, the lowest-lying excited state of ^{16}O is of a $1p$-$1h$ character. We can therefore rule out α-cluster correlations. Such states appear at higher excitation energies in our calculations. In Ref. [38], we obtained converged results for the lowest-lying 3^-_1 state of ^{16}O. For the N^{3}LO interaction, we have an excitation energy of about 12 MeV, almost 6 MeV above the experimental value of 6.13 MeV. We have checked in $N = 7$ shells that the excitation energy changes from 12.61 at $\hbar\Omega = 10$ MeV to 12.5 MeV at $\hbar\Omega = 14$ MeV. The excited states of ^{16}O we have studied are almost independent of the oscillator energy at $N = 8$ shells.

The low-lying excited states of ^{16}O and, in general, states that involve cross-shell excitations have always eluded a proper microscopic description (see, for example, Refs. [123–128] and references therein). Let us concentrate on the lowest-energy 3^-_1 state of ^{16}O. In a zero-order approximation, this state may be regarded as a state that arises from the single $i \to a$ excitation from the $i = 0p_{1/2}$ hole state to the $a = 0d_{5/2}$ particle state. Relative to the ^{16}O ground state, the energy required to produce such an excitation equals

$$\begin{aligned}\Delta\epsilon_\pi &= \epsilon_\pi(0d_{5/2}) - \epsilon_\pi(0p_{1/2}) \\ &= [\mathrm{BE}(^{16}\mathrm{O}) - \mathrm{BE}(^{17}\mathrm{F})] + [\mathrm{BE}(^{16}\mathrm{O}) - \mathrm{BE}(^{15}\mathrm{N})] \\ &= 11.526\,\mathrm{MeV}, \end{aligned} \tag{42}$$

for the proton case, and

$$\begin{aligned}\Delta\epsilon_\nu &= \epsilon_\nu(0d_{5/2}) - \epsilon_\nu(0p_{1/2}) \\ &= [\mathrm{BE}(^{16}\mathrm{O}) - \mathrm{BE}(^{17}\mathrm{O})] + [\mathrm{BE}(^{16}\mathrm{O}) - \mathrm{BE}(^{15}\mathrm{O})] \\ &= 11.521\,\mathrm{MeV}, \end{aligned} \tag{43}$$

for the neutron case, where the acronym BE in the above equations represents the relevant total binding energies. In calculating the above values of the $1p$-$1h$ excitation energies $\Delta\epsilon_\pi$ and $\Delta\epsilon_\nu$ that provide us with the zero-order estimates of the excitation energy of the lowest 3^-_1 state of ^{16}O, we used the experimental binding energies listed in Table I. As we can see from Eqs. (42) and (43), the proton and neutron excitation energies are nearly identical. This reflects a well-known feature of spin-isospin saturated systems. Without interactions among nucleons and with the above single-particle orbits used as the only active degrees of freedom, all negative parity states with quantum numbers $J^\pi = 2^-, 3^-$ would be at the above energies of approximately 11.5 MeV. The interactions among nucleons lower the energy of the first excited 3^- state by $11.5 - 6.1 = 5.4$ MeV.

Let us now compare the approximate energy spacing defining the lowest 3^- state of ^{16}O, resulting from the use of experimental binding energies, as shown above (11.5 MeV), with the values of $\Delta\epsilon_\pi$ and $\Delta\epsilon_\nu$ based on the results of coupled-cluster calculations for the binding energies of ^{16}O and valence systems around ^{16}O obtained with the N^{3}LO interaction and eight major oscillator shells. These results are $\Delta\epsilon_\pi = 15.846$ MeV and $\Delta\epsilon_\nu = 15.789$ MeV, for proton and neutron excitations, respectively, with almost the same difference between the proton and neutron cases as observed in experiment. The authors of Ref. [110] obtained 14.72 and 14.64 MeV for protons and neutrons, respectively, using the same N^{3}LO interaction as used here. Using the above elementary picture of the $1p$-$1h$ excitation defining the lowest 3^- state of ^{16}O, which involves only two orbits in the definition of the relevant model space, we can see that we

are off by approximately $15.8 - 11.5 = 4.3$ MeV, when we compare the $\Delta\epsilon_\pi$ and $\Delta\epsilon_\nu$ energy spacings resulting from coupled-cluster calculations with the experimental estimates of these spacings. This difference is obviously an interaction- and method-dependent result. It is, however, converged as a function of the number of oscillator shells in a basis set, showing that the discrepancy of 4.3 MeV between theory and experiment for the energy gap between the $0p$ and $1s0d$ shells accounts for a large fraction of the missing 6 MeV needed to reproduce the first 3^- state of ^{16}O. This is, perhaps, the most likely candidate for a consistent explanation of the large difference between converged coupled-cluster result for the lowest 3^- state of ^{16}O and experiment reported in Ref. [38]. This conclusion is not substantially altered by the slight $\hbar\Omega$ dependence within the calculations.

The above analysis indicates that a large fraction of the difference between theory and experiment can be traced in this case to errors in reproducing the experimental binding energies of ^{16}O and valence systems around ^{16}O by coupled-cluster methods employing pairwise interactions only. This allows us to conclude that a 6 MeV difference between coupled-cluster result and experiment for the lowest 3^- state of ^{16}O is primarily caused by the lack of three-body interactions in our calculations and much less by the approximate treatment of particle correlations by the coupled-cluster methods used in our studies. The above analysis also implies that with an adjusted gap between the $0p$ and $1s0d$ shells, one should be able to get a better reproduction of the excited states of ^{16}O which have a well-defined $1p$-$1h$ structure, such as the lowest 3^- state discussed here. One possible strategy for describing excited states of closed-shell nuclei dominated by $1p$-$1h$ excitations might be to keep the original two-body Hamiltonian and add additional three-body terms via corrections to the single-particle energies, as advocated recently by Zuker [129,130].

C. Comparisons among different interaction models

The binding energies per particle for the three interaction models examined in this work, namely N^{3}LO, CD-Bonn, and V_{18}, are listed in Table VI. We show only the results obtained with eight major oscillator shells, because convergence patterns with the number of major oscillator shells that characterize the N^{3}LO, CD-Bonn, and V_{18} interactions are practically identical.

As expected, the CD-Bonn interaction gives more attraction than N^{3}LO, whereas the Argonne V_{18} interaction model yields less attraction than the other two models. The CD-Bonn potential has the weakest tensor force of the three interactions studied here, whereas the V_{18} interaction has the strongest tensor force component. It is well-known that an interaction model with a weak tensor force yields less quenching in the medium for the important $^3S_1$ and $^3D_1$ partial wave contributions to various matrix elements of the Hamiltonian. The quenching is ascribed to both a Pauli effect and an energy dependence reflected in second- and higher-order terms (see, for example, Ref. [6] for a discussion of this topic in both nuclei and nuclear matter). Although all interaction models fit

TABLE VI. A comparison of the binding energies per particle for ^{15}O and ^{15}N (the PR-EOMCCSD values), ^{16}O (the CCSD values), and ^{17}O and ^{17}F (the PA-EOMCCSD values), obtained with the N^{3}LO [31], CD-Bonn [29], and V_{18} [28] potentials, and eight major oscillator shells, with the experimental data taken from Ref. [117]. All entries are in MeV. For the CD-Bonn and N^{3}LO interactions, we used $\hbar\Omega = 11$ MeV. For V_{18}, we used $\hbar\Omega = 10$ MeV. For eight major shells $\beta_{\mathrm{CoM}} = 0.0$.

Nucleus	Interaction			Expt.
	N^{3}LO	CD-Bonn	V_{18}	
^{15}O	6.643	7.584	5.246	7.464
^{15}N	6.824	7.751	5.414	7.699
^{16}O	7.406	8.327	5.897	7.976
^{17}O	7.150	8.032	5.617	7.751
^{17}F	6.987	7.879	5.462	7.542

properties of the deuteron and the scattering data with a χ^2 per datum close to 1, the nonlocalities that are introduced because of the way the interactions are constructed are responsible for different results in a many-body context. Indeed, the N^{3}LO and CD-Bonn models are nonlocal interactions defined in momentum space. While the N^{3}LO model is based on chiral Lagrangians with nucleons and pions as degrees of freedom, including the noniterative 2π diagrams at chiral fourth order, the CD-Bonn interaction is a traditional meson-exchange model that includes the six low-mass mesons $\pi, \delta, \rho, \Omega, \eta$ and the fictitious σ meson, which is a 2π resonance. The Argonne V_{18} model is based on a local r-space parametrization, dominated by one-pion exchange. The strength of the nuclear tensor force is intimately connected with the nonlocalities of the different nucleon-nucleon forces. Depending on how it is quenched in a many-body context, one may get less or more attraction. The attractive part of, for example, the $^3S_1$ partial wave contribution is more attractive in the medium for an interaction with a weak tensor than for one with a strong tensor force. Such features are clearly seen in the coupled-cluster results reported in Table VI, where the potential with the weakest tensor force, CD-Bonn, yields more binding than the two other models. Our results for the CD-Bonn interaction show more binding than the calculations of Fujii *et al.* [110].

Based on our earlier work [38], the triply excited clusters and the related $3p$-$2h$ and $3h$-$2p$ excitations in the particle-attaching and particle-removing $R_\mu^{(A+1)}$ and $R_\mu^{(A-1)}$ operators of the PA-EOMCC and PR-EOMCC theories are expected to have little impact on the calculated binding energies. Horoi *et al.* [131] show that for ^{4}He and up to seven major shells, the coupled-cluster method agrees excellently (to within 0.1 MeV; often much better) with shell-model calculations and that corrections to the total binding energy due to T_3 clusters are on the order of 1 MeV or less (see Ref. [37] for similar findings for smaller basis sets). Furthermore, the results with a G-matrix applied to a shell-model calculation with eight major shells exhibit a vanishing starting energy dependence [131]. We claim therefore that, except for a small correction due to triples and a weak starting energy dependence [38], the lack of agreement between coupled-cluster and experimental binding

energies is primarily because of the missing physics in our Hamiltonians.

The main conclusion that one can derive from the results of our coupled-cluster calculations with different interactions is that every nucleon-nucleon interaction model needs its own three-body potential. The Argonne group has derived sophisticated three-body interaction terms (see, for example, the extensive elaboration of Ref. [132]). The parameters entering their three-body interaction models are fitted to reproduce properties of light nuclei. These three-body terms follow much of the same pion-exchange picture adopted in the construction of the Argonne V_{18} interaction. For the CD-Bonn interaction one would need to derive three-body terms based on a meson-exchange picture, as outlined, for example, by the Bochum group [133]. However, no such model that accompanies this interaction has been fully developed. The situation for models based on effective field theory is much better as three-body terms arise quite naturally at given orders in the expansion parameter [120]. Our coupled-cluster results indicate that every interaction, because of different nonlocalities, has its own three-body component reflected in different binding energies and different spin-orbit splittings [the $(3/2)^-_1 - (1/2)^-_1$ spacings in ^{15}O and ^{15}N and the $(3/2)^+_1 - (5/2)^+_1$ spacings in ^{17}O and ^{17}F], as demonstrated in Table VI, which lists binding energies per nucleon, and Table VII, which lists the corresponding low-lying excited states of the valence systems around ^{16}O examined in this work.

As shown in Table VII, the CD-Bonn and the N^{3}LO models result in the largest spin-orbit splittings (much larger than in the case of V_{18}). To examine this behavior in some detail, we have computed all diagrams through third order in the G matrix for $\hbar\Omega = 14$ MeV, using many-body perturbation theory as described in Ref. [6], including folded diagrams to infinite order. For example, at the Hartree-Fock level, which corresponds to the first order in the G matrix, the spin-orbit splittings for neutrons between the two hole states in the $0p$ shell are 4.85, 4.41, and 3.91 MeV for the CD-Bonn, N^{3}LO, and V_{18} interaction models, respectively. Because we are dealing with spin-isospin saturated systems, the results for protons are almost the same. The Hartree-Fock term yields the largest contribution and receives important contributions from the short-range two-body spin-orbit force. However, there is also a considerable contribution to the splitting that originates from the second-order $2h$-$1p$ and $2p$-$1h$ terms. The corresponding second-order contributions are 1.81, 1.73, and 1.35 MeV for the same three interactions, respectively. These perturbation theory estimates agree with the ways the $(3/2)^-_1 - (1/2)^-_1$ spacings in ^{15}O and ^{15}N and the $(3/2)^+_1 - (5/2)^+_1$ spacings in ^{17}O and ^{17}F, obtained in the corresponding PR-EOMCCSD and PA-EOMCCSD calculations, vary with the interaction. This analysis illustrates, at least to some extent, the role played by the quenching of the tensor force via the second- and higher-order terms in many-body perturbation theory in different interaction models. The perturbative results do not stabilize, however, as functions of the oscillator energy, a result that is in close agreement with the findings reported by Fujii *et al.* [110]. With increasing $\hbar\Omega$, the single-particle splittings increase if one uses an unperturbed harmonic oscillator basis. This is mostly because of the way we treat the Coulomb interaction, as discussed above.

It is interesting to note that despite the apparent differences between the coupled-cluster results obtained with different pairwise interaction models, the relative binding energies of ^{15}O, ^{15}N, ^{16}O, ^{17}O, and ^{17}F obtained with different interactions are in good agreement with experiment and with each other. For example, as already mentioned the difference between experimental binding energies of ^{16}O and ^{17}O is 0.225 MeV per particle. The CCSD and PA-EOMCCSD ground-state energies in Table VI of ^{16}O and ^{17}O resulting from the calculations with eight major oscillator shells differ by 0.256 MeV per particle for N^{3}LO, 0.295 MeV per particle for CD-Bonn, and 0.280 MeV per particle for V_{18}. Similarly, the difference between experimental binding energies of ^{16}O and ^{15}O is 0.512 MeV per particle, whereas the CCSD and PR-EOMCCSD ground-state energies of ^{16}O and ^{15}O differ by 0.763, 0.743, and 0.651 MeV per particle for the N^{3}LO, CD-Bonn, and V_{18} potentials, respectively. Here the differences with experiment are somewhat greater than in the case of ^{16}O and ^{17}O, but the overall agreement among different potentials is still good. The differences between the binding energies for the $A = 15$ nuclei and for the $A = 17$ nuclei obtained with different interactions are close to one another and to the experimental values, too. According to Table VI, the experimental value of the binding energy difference BE(^{15}N)−BE(^{15}O) is 0.235 MeV per particle. The PR-EOMCCSD calculations with the N^{3}LO, CD-Bonn, and V_{18} interactions give 0.181, 0.167, and 0.168 MeV per particle, respectively, for the same binding energy difference. Similarly, the experimental value of the binding energy difference BE(^{17}O)−BE(^{17}F) is 0.209 MeV per particle. The PA-EOMCCSD calculations with the N^{3}LO, CD-Bonn, and V_{18} potentials give 0.163, 0.153, and 0.155 MeV per particle, respectively, for the same binding energy difference. Despite the substantial differences between binding energies resulting from the calculations with different interactions, which are likely affected by the three-body forces

TABLE VII. A comparison of the energies of the low-lying excited states of ^{15}O, ^{15}N, ^{17}O, and ^{17}F, relative to the corresponding ground-state energies [the $(1/2)^-_1$ states of ^{15}O and ^{15}N and the $(5/2)^+_1$ states of ^{17}O and ^{17}F], obtained with the PR-EOMCCSD (^{15}O and ^{15}N) and PA-EOMCCSD (^{17}O and ^{17}F) methods, the N^{3}LO [31], CD-Bonn [29], and V_{18} [28] potentials, and eight major oscillator shells, with the experimental data taken from Ref. [121]. All entries are in MeV. For the CD-Bonn and N^{3}LO interactions, we used $\hbar\Omega = 11$ MeV. For V_{18}, we used $\hbar\Omega = 10$ MeV. For eight major shells $\beta_{\mathrm{CoM}} = 0.0$.

Excited state	Interaction			Expt.
	N^{3}LO	CD-Bonn	V_{18}	
^{15}O $(3/2)^-_1$	6.264	7.351	4.452	6.176
^{15}N $(3/2)^-_1$	6.318	7.443	4.499	6.323
^{17}O $(3/2)^+_1$	5.675	6.406	3.946	5.084
^{17}O $(1/2)^+_1$	−0.025	0.311	−0.390	0.870
^{17}F $(3/2)^+_1$	5.891	6.677	4.163	5.000
^{17}F $(1/2)^+_1$	0.428	0.805	0.062	0.495

that are expected to be different for different pairwise interactions, the binding energies per nucleon resulting from our PR-EOMCCSD/CCSD/PA-EOMCCSD calculations with eight major oscillator shells satisfy $^{15}O < ^{15}N < ^{17}F < ^{17}O < ^{16}O$, independent of the interaction used in coupled-cluster calculations.

IV. CONCLUSIONS AND PERSPECTIVES

We summarize here our main conclusions and perspectives for future studies.

(i) To our knowledge, this is the first application of the *ab initio* coupled-cluster theory employing the renormalized form of the Hamiltonian, combined with the PA-EOMCC and PR-EOMCC formalisms for open-shell many-fermion systems, to nuclear valence systems with one valence particle or one valence hole. We have shown that one can obtain reasonably well converged results with given two-body Hamiltonians for both binding energies and at least some low-lying excited states. The systems whose properties have been studied in this work were ^{15}O, ^{15}N, ^{17}O, and ^{17}F. An emphasis has been placed on states dominated by one-quasiparticle configurations. The discrepancies between the results of large-scale coupled-cluster calculations for these nuclei and the corresponding experimental data for such states have been traced to the Hamiltonians used in the calculations and much less to the correlations neglected in the coupled-cluster approximations employed in this study, such as triples corrections. These corrections are, however, small, as demonstrated in comparisons with shell-model calculations using the same Hamiltonian [37,131]. The calculations of Horoi *et al.* [131] demonstrate also that the starting energy dependence is very weak for eight major shells. For the excited valence particle states there is, however, experimental and theoretical indication that these states may couple to more complicated correlations than those included at the PA-EOMCCSD level. Furthermore, while our ground-state energies stabilize as functions of the size of the model space and the chosen oscillator energy, our Coulomb energy differences depend still on the oscillator energy.

(ii) Three different nucleon-nucleon interactions have been used to define our two-body Hamiltonians. These are the N^{3}LO model [31], the CD-Bonn interaction [29], and the V_{18} model of the Argonne group [28]. All of these interactions yield different binding energies and different energies of the excited states. The different binding energies and spin-orbit splittings can be related to varying nonlocalities in the nucleon-nucleon interactions. Of particular interest here has been the role played by the nuclear tensor force. The different behavior of the three interaction models examined in this study points to the need for the development of interaction specific three-body forces.

(iii) We have also demonstrated that most of the discrepancy between theory and experiment for the $1p$-$1h$ negative parity states in ^{16}O, including the lowest 3_1^- state examined in our earlier work [38], can be retraced to the difference between the theoretical and experimental values of the relevant energy gaps between neutron or proton states in the $0p$ and $1s0d$ shells.

(iv) Despite the differences among interactions, the relative binding energies of the ^{15}O,^{15}N,^{17}F,^{17}O, and ^{16}O resulting from the coupled-cluster calculations seem to be independent of the interaction and in good agreement with experiment. The $(3/2)_1^- - (1/2)_1^-$ spacings in ^{15}O and ^{15}N resulting from the converged coupled-cluster calculations with the N^{3}LO interaction are in good agreement with experiment, indicating that the spin-orbit force associated with an eventual three-body force for N^{3}LO should be small.

There are several obvious extensions to this work. First of all, the need for an inclusion of three-body interactions sets the agenda for forthcoming studies. Moreover, our Coulomb energy differences need further studies and it may be useful to examine the role of $3p$-$2h$ and $3h$-$2p$ correlations in the PA-EOMCC and PR-EOMCC calculations, which we neglected in this study. For the hole states considered here, $3h$-$2p$ correlations are expected to be small, because the states of ^{15}O and ^{15}N that we have examined show relatively small departures from an independent-particle picture and because the underlying T_3 cluster contributions that define the reference ^{16}O system are small [37,38,131]. However, the $3p$-$2h$ correlations may be important for the excited valence particles of ^{17}F and ^{17}O. There we have also tacitly assumed that the $0d_{3/2}$ states of ^{17}F and ^{17}O are bound states. These states are resonances, and it is not yet entirely clear how the nonresonant continuum may affect the description of these states. The inclusion of such contributions in the description of these states is another important point to explore, as demonstrated in the recent works on the Gamow shell-model and complex-scaling techniques [134–136].

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APPENDIX: FACTORIZED FORM OF THE CCSD, PA-EOMCCSD, AND PR-EOMCCSD EQUATIONS

In this appendix, we present the working equations defining the CCSD, PA-EOMCCSD, and PR-EOMCCSD methods exploited in this study. All of the equations are expressed in terms of the one- and two-body matrix elements of the Hamiltonian in the normal-ordered form, f_α^β and $v_{\alpha\beta}^{\gamma\delta}$, respectively [cf. Eq. (5); in our case, f_α^β and $v_{\alpha\beta}^{\gamma\delta}$ are the one- and two-body matrix elements of the normal-ordered form of the effective Hamiltonian H, Eq. (4)], the t_a^i and t_{ab}^{ij} cluster amplitudes defining the underlying A-particle ground-state CCSD problem, and, in the case of the PA-EOMCCSD and PR-EOMCCSD approaches, the r_a, r_{ab}^j, r^i, and r_b^{ij} amplitudes defining the particle-attaching (r_a and r_{ab}^j) and particle-removing (r^i and r_b^{ij}) operators, $R_\mu^{(A+1)}(2p\text{-}1h)$ and $R_\mu^{(A-1)}(2h\text{-}1p)$, Eqs. (33) and (34), respectively. As explained in Sec. II D, the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations can be cast into a computationally efficient factorized form expressed in terms of the one- and two-body matrix elements of the CCSD similarity-transformed Hamiltonian $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$, $\bar{h}_\alpha^\beta$ and $\bar{h}_{\alpha\beta}^{\gamma\delta}$, respectively, and a few additional intermediates that are generated in a recursive manner. The complete set of one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$ and other intermediates that are needed to set up the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations is given in Table VIII.

The ground-state CCSD equations for the singly and doubly excited cluster amplitudes t_a^i and t_{ab}^{ij}, Eqs. (24) and (25), can be given the following, computationally efficient form:

$$\bar{h}_a^i \equiv f_a^i + I_a^{\prime e} t_e^i - \bar{h}_m^i t_a^m - v_{ma}^{ie} t_e^m + \bar{h}_m^e t_{ea}^{mi} - \tfrac{1}{2}\bar{h}_{mn}^{ie} t_{ae}^{mn} + \tfrac{1}{2} v_{am}^{ef} t_{ef}^{im} = 0, \tag{A1}$$

$$\bar{h}_{ab}^{ij} \equiv v_{ab}^{ij} + \mathcal{A}_{ab}\mathcal{A}^{ij}\Big[\tfrac{1}{2} I_{ab}^{ie} t_e^j - \tfrac{1}{2} I_{mb}^{ij} t_a^m + \tfrac{1}{2} I_b^e t_{ae}^{ij} + \tfrac{1}{8} v_{ab}^{ef} t_{ef}^{ij} + \tfrac{1}{8}\bar{h}_{mn}^{ij} t_{ab}^{mn} - I_{mb}^{ie} t_{ae}^{mj} - \tfrac{1}{2}\bar{h}_m^j t_{ab}^{im}\Big] = 0. \tag{A2}$$

Note that the left-hand sides of Eqs. (24) and (25) [or Eqs. (A1) and (A2)] represent, respectively, the one- and two-body matrix elements $\bar{h}_a^i$ and $\bar{h}_{ab}^{ij}$ of $\bar{H}_N(\mathrm{CCSD})$ [see Eqs. (15) and (16)]. The relevant intermediates can be found in Table VIII. The antisymmetrizers $\mathcal{A}_{pq} = \mathcal{A}^{pq}$, which enter Eq. (A2) and other equations presented in this Appendix, are defined as

$$\mathcal{A}_{pq} \equiv \mathcal{A}^{pq} = 1 - (pq), \tag{A3}$$

with (pq) representing a transposition of two indices. Once the above equations are solved for t_a^i and t_{ab}^{ij}, the ground-state CCSD energy is calculated using the formula [cf. Eq. (23)]

$$E_0^{(A)}(M) = \langle\Phi|H|\Phi\rangle + f_i^a t_a^i + \tfrac{1}{4} v_{ij}^{ab}\big(t_{ab}^{ij} + 2 t_a^i t_b^j\big), \tag{A4}$$

which is valid for any truncation scheme $M \geqslant 2$.

Once the t_a^i and t_{ab}^{ij} amplitudes are determined and the ground-state CCSD energy of the reference A-body system is known, we can set up and solve the eigenvalue equations defining the PA-EOMCCSD and PR-EOMCCSD methods. The PA-EOMCCSD equations for the energy differences $\omega_\mu^{(A+1)} = E_\mu^{(A+1)} - E_0^{(A)}$ and the $1p$ and $2p$-$1h$ amplitudes, r_a and r_{ab}^j, respectively, defining the ground and excited states of the $(A+1)$-particle system, can be given the following,

TABLE VIII. Explicit algebraic expressions for the one- and two-body matrix elements of $\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD})$($\bar{h}_\alpha^\beta$ and $\bar{h}_{\alpha\beta}^{\gamma\delta}$, respectively) and other intermediates (designated by I) used to construct the computationally efficient form of the CCSD, PA-EOMCCSD, and PR-EOMCCSD equations.

Intermediate	Expression[a]
$\bar{h}_i^a$	$f_i^a + v_{im}^{ae} t_e^m$
$\bar{h}_i^j$	$f_i^j + v_{im}^{je} t_e^m + \tfrac{1}{2} v_{mi}^{ef} t_{ef}^{mj} + \bar{h}_i^e t_e^j$
$\bar{h}_a^b$	$I_a^b - \bar{h}_m^b t_a^m$
$\bar{h}_{ai}^{bc}$	$v_{ai}^{bc} - v_{mi}^{bc} t_a^m$
$\bar{h}_{ij}^{ka}$	$v_{ij}^{ka} + v_{ij}^{ea} t_e^k$
$\bar{h}_{ab}^{cd}$	$v_{ab}^{cd} + \tfrac{1}{2} v_{mn}^{cd} t_{ab}^{mn} - \bar{h}_{am}^{cd} t_b^m + v_{bm}^{cd} t_a^m$
$\bar{h}_{ij}^{kl}$	$v_{ij}^{kl} + \tfrac{1}{2} v_{ij}^{ef} t_{ef}^{kl} - \bar{h}_{ij}^{le} t_e^k + v_{ij}^{ke} t_e^l$
$\bar{h}_{ia}^{jb}$	$I_{ia}^{\prime jb} - v_{im}^{eb} t_{ea}^{jm} - \bar{h}_{im}^{jb} t_a^m$
$\bar{h}_{ab}^{ic}$	$v_{ab}^{ic} + v_{ab}^{ec} t_e^i - \bar{h}_{mb}^{ic} t_a^m + I_{ma}^{\prime ic} t_b^m$
	$-\bar{h}_m^c t_{ab}^{im} + \bar{h}_{bm}^{ce} t_{ae}^{im} - v_{am}^{ce} t_{be}^{im} + \tfrac{1}{2}\bar{h}_{nm}^{ic} t_{ab}^{nm}$
$\bar{h}_{ia}^{jk}$	$v_{ia}^{jk} + \bar{h}_{mi}^{jk} t_a^m - v_{ia}^{ke} t_e^j + \mathcal{A}^{jk}\bar{h}_{im}^{je} t_{ae}^{km}$
	$+\bar{h}_i^e t_{ea}^{jk} + I_{ia}^{\prime je} t_e^k - \tfrac{1}{2} v_{ai}^{ef} t_{ef}^{jk}$
$I_a^{\prime b}$	$f_a^b + v_{am}^{be} t_e^m$
I_a^b	$I_a^{\prime b} - \tfrac{1}{2} v_{mn}^{eb} t_{ea}^{mn}$
I_{ab}^{ic}	$v_{ab}^{ic} + v_{ab}^{ec} t_e^i + v_{mb}^{ec} t_{ae}^{im}$
I_{ia}^{jk}	$\bar{h}_{ia}^{jk} - \tfrac{1}{2}\bar{h}_{mi}^{jk} t_a^m$
I_{ia}^{jb}	$-\tfrac{1}{2} v_{im}^{eb} t_{ea}^{jm}$
$I_{ia}^{\prime jb}$	$v_{ia}^{jb} + v_{ia}^{eb} t_e^j$
I_m	$\tfrac{1}{2} v_{mn}^{ef} r_{ef}^n$
I^e	$-\tfrac{1}{2} v_{mn}^{ef} r_f^{mn}$

[a] Summation over repeated upper and lower indices is assumed. $f_\alpha^\beta = \langle\alpha|f|\beta\rangle$ and $v_{\alpha\beta}^{\gamma\delta} = \langle\alpha\beta|v|\gamma\delta\rangle - \langle\alpha\beta|v|\delta\gamma\rangle$ are the one- and two-body matrix elements of the Hamiltonian in the normal-ordered form, Eq. (5) and the t_a^i and t_{ab}^{ij} are the singly and doubly excited cluster amplitudes defining the ground-state CCSD wave function of the A-body reference system.

computationally efficient, form:

$$\langle \Phi^a | \big[\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) R_\mu^{(A+1)}(2p\text{-}1h) \big]_C | \Phi \rangle = \bar{h}_a^e r_e + \bar{h}_m^e r_{ae}^m + \tfrac{1}{2} \bar{h}_{am}^{ef} r_{ef}^m = \omega_\mu^{(A+1)} r_a, \quad \text{(A5)}$$

$$\langle \Phi_j^{ab} | \big[\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) R_\mu^{(A+1)}(2p\text{-}1h) \big]_C | \Phi \rangle = \mathcal{A}_{ab} \big[-\tfrac{1}{2} \bar{h}_{ab}^{je} r_e + \bar{h}_a^e r_{eb}^j - \tfrac{1}{2} \bar{h}_m^j r_{ab}^m + \tfrac{1}{4} \bar{h}_{ab}^{ef} r_{ef}^{\ j} - \bar{h}_{ma}^{je} r_{eb}^m - \tfrac{1}{2} I_m t_{ab}^{mj} \big] = \omega_\mu^{(A+1)} r_{ab}^{\ j}. \quad \text{(A6)}$$

Similarly, we can use the CCSD values of the singly and doubly excited cluster amplitudes defining the ground-state wave function of the reference A-body system to set up the PR-EOMCCSD eigenvalue equations for the energy differences $\omega_\mu^{(A-1)} = E_\mu^{(A-1)} - E_0^{(A)}$ and the $1h$ and $2h$-$1p$ amplitudes, r^i and r_b^{ij}, respectively, defining the ground and excited states of the $(A-1)$-particle system. The computationally efficient form of the PR-EOMCCSD equations is as follows:

$$\langle \Phi_i | \big[\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) R_\mu^{(A-1)}(2h\text{-}1p) \big]_C | \Phi \rangle = -\bar{h}_m^i r^m + \bar{h}_m^e r_e^{im} - \tfrac{1}{2} \bar{h}_{mn}^{ie} r_e^{mn} = \omega_\mu^{(A-1)} r^i, \quad \text{(A7)}$$

$$\langle \Phi_{ij}^{\ b} | \big[\bar{H}_{N,\mathrm{open}}(\mathrm{CCSD}) R_\mu^{(A-1)}(2h\text{-}1p) \big]_C | \Phi \rangle = A^{ij} \big[-\tfrac{1}{2} \bar{h}_{mb}^{ij} r^m - \bar{h}_m^i r_b^{mj} + \tfrac{1}{2} \bar{h}_b^e r_e^{ij} + \tfrac{1}{4} \bar{h}_{mn}^{ij} r_b^{mn} - \bar{h}_{mb}^{ie} r_e^{mj} + \tfrac{1}{2} I^e t_{eb}^{ij} \big] = \omega_\mu^{(A-1)} r_b^{ij}. \quad \text{(A8)}$$

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