Ab Initio Calculations of Even Oxygen Isotopes with Chiral Two- Plus Three-Nucleon Interactions

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(Dated: March 15, 2013)

We formulate the In-Medium Similarity Renormalization Group (IM-SRG) for open-shell nuclei using a multi-reference formalism based on a generalized Wick theorem introduced in quantum chemistry. The resulting multi-reference IM-SRG (MR-IM-SRG) is used to perform the first *ab initio* study of even oxygen isotopes with chiral NN and 3N Hamiltonians, from the proton to the neutron drip lines. We obtain an excellent reproduction of experimental ground-state energies with quantified uncertainties, which is validated by results from the Importance-Truncated No-Core Shell Model and the Coupled Cluster method. The agreement between conceptually different many-body approaches and experiment highlights the predictive power of current chiral two- and three-nucleon interactions, and establishes the MR-IM-SRG as a promising new tool for *ab initio* calculations of medium-mass nuclei far from shell closures.

Introduction. Neutron-rich nuclei are the focus of the experimental program of current and next-generation rare isotope facilities. Emerging phenomena such as halos or neutron skins make such nuclei ideal laboratories to study nuclear interactions in delicately tuned scenarios. These phenomena are driven by the evolution of shell structure along isotopic or isotonic chains, and motivate the use of *ab initio* many-body calculations to provide a description from first principles. Such calculations make it possible to confront modern nuclear Hamiltonians from chiral effective field theory (EFT) [1, 2] with a wealth of data beyond few-body systems.

For s- and p-shell nuclei, the *ab initio* No-Core Shell Model (NCSM) [3, 4] provides the necessary capabilities for studies of isotopic chains, but for medium-mass nuclei this approach is not feasible because of its large computational effort. Many-body techniques with more modest computational scaling, such as the Coupled Cluster (CC) method, can be used to probe nuclei in the vicinity of shell closures [5–7], but are usually not applicable for open-shell nuclei far from shell closures. For such nuclei, a self-consistent Gor'kov formalism was developed recently [8, 9], but at the present stage this approach is limited to second-order terms in the many-body perturbation expansion.

In this Letter, we describe the extension of the In-Medium Similarity Renormalization Group (IM-SRG) framework of Refs. [10, 11] to open-shell nuclei by means of a multi-reference formulation. We use the resulting MR-IM-SRG in conjunction with two other many-body approaches, the Importance-Truncated No-Core Shell Model (IT-NCSM) and the CC method, to perform the first *ab initio* study of even oxygen isotopes with chiral NN+3N Hamiltonians.

Formalism. The main tools for the derivation of the MR-IM-SRG are the generalized normal-ordering and Wick theorem introduced by Kutzelnigg and Mukherjee [12]. Let us denote a string of creation and annihilation operators in tensorial form,

$$A^{1...k}_{l...N} \equiv a^\dagger_1 \ldots a^\dagger_k a_N \ldots a_l \,, \tag{1}$$

and expand it in terms of components that are normal-ordered with respect to a given reference state $|\Phi\rangle$, which can be a superposition of Slater determinants in the multi-reference case [13]. We obtain

$$\begin{aligned} A^{1...k}_{l...N} =\; & :A^{1...k}_{l...N}: -\lambda^1_l :A^{23...k}_{mn...N}: -\lambda^1_m :A^{23...k}_{ln...N}: +\ldots \\ & + (\lambda^1_l\lambda^2_m - \lambda^1_m\lambda^2_l + \lambda^{12}_{lm}) :A^{3...k}_{n...N}: +\ldots\,, \end{aligned} \tag{2}$$

where we have introduced irreducible one- and two-body density matrices $\lambda^{(1)}$ and $\lambda^{(2)}$:

$$\lambda^1_2 \equiv \langle\Phi|A^1_2|\Phi\rangle\,, \quad \lambda^{12}_{34} \equiv \langle\Phi|A^{12}_{34}|\Phi\rangle - \lambda^1_2\lambda^3_4 + \lambda^1_3\lambda^2_4\,. \tag{3}$$

The particle rank of the irreducible density matrices is evident from the single-particle indices. Generally, up to n-body irreducible density matrices $\lambda^{(n)}$ appear in the expansion of an n-body operator, which are defined recursively in terms of density matrices of lower rank and encode information about n-body correlations in the reference state [12]. For an independent-particle state, all of these matrices except $\lambda^{(1)}$ vanish identically.

Products of normal-ordered operators can be expanded by means of a generalized Wick theorem (GWT), e.g.,

$$\begin{aligned} & :A^{12}_{56}::A^{34}_{78}: \\ = & :A^{1234}_{5678}: +\lambda^1_7 :A^{234}_{568}: -\xi^3_5 :A^{124}_{678}: +\ldots \\ & + \left(\lambda^1_7\lambda^2_8 - \lambda^1_8\lambda^2_7 + \lambda^{12}_{78}\right):A^{34}_{56}: -\lambda^{12}_{57}:A^{34}_{68}: +\ldots\,, \end{aligned} \tag{4}$$

where $\xi^1_2 \equiv \lambda^1_2 - \delta^1_2$ [14]. In addition to simple contractions containing $\lambda^{(1)}$ and $\xi^{(1)}$ which also occur in the standard Wick theorem, we obtain terms involving $\lambda^{(2)}, \ldots, \lambda^{(n)}$. Each density matrix must have at least one index from each of the operators in the product — other terms vanish due to the initial normal-ordering (2), which greatly simplifies the algebra [14]. In the following, we work in natural orbitals, i.e., the eigenbasis of $\lambda^{(1)}$, where

$$\lambda^1_2 = n_1\delta^1_2\,, \quad \xi^1_2 = -\bar{n}_1\delta^1_2 \equiv -(1-n_1)\delta^1_2\,, \tag{5}$$

and the eigenvalues are the occupation numbers $0 \le n_a \le 1$.

We now consider the IM-SRG operator flow equation

$$\frac{d}{ds}H(s) = [\eta(s), H(s)],\qquad(6)$$

which is used to solve the many-body problem by a continuous unitary transformation that decouples the ground-state of the Hamiltonian $H(s)$ from excitations [10, 11]. Suppressing the flow parameter s for brevity, we apply the generalized normal-ordering to H and the generator η, and evaluate the commutator using the GWT to obtain the zero-, one-, and two-body MR-IM-SRG flow equations:

$$\frac{dE}{ds} = \sum_{ab}(n_a - n_b)\left(\eta_b^a f_a^b - f_b^a \eta_a^b\right)$$
$$+ \frac{1}{4}\sum_{abcd}\left(\eta_{cd}^{ab}\Gamma_{ab}^{cd} - \Gamma_{cd}^{ab}\eta_{ab}^{cd}\right)n_a n_b \bar{n}_c \bar{n}_d$$
$$+ \frac{1}{4}\sum_{abcd}\left(\frac{d}{ds}\Gamma_{cd}^{ab}\right)\lambda_{cd}^{ab},\qquad(7)$$

$$\frac{d}{ds}f_2^1 = \sum_a \eta_a^1 f_2^a + \sum_{ab}\eta_b^a \Gamma_{a2}^{b1}(n_a - n_b)$$
$$+ \frac{1}{2}\sum_{abc}\eta_{bc}^{1a}\Gamma_{2a}^{bc}\left(n_a\bar{n}_b\bar{n}_c + \bar{n}_a n_b n_c\right)$$
$$+ \frac{1}{4}\sum_{abcde}\eta_{bc}^{1a}\Gamma_{2a}^{de}\lambda_{bc}^{de} + \sum_{abcde}\eta_{bc}^{1a}\Gamma_{2d}^{be}\lambda_{cd}^{ae}$$
$$- \frac{1}{2}\sum_{abcde}\left(\eta_{2b}^{1a}\Gamma_{ae}^{cd}\lambda_{be}^{cd} - \eta_{2b}^{1a}\Gamma_{de}^{bc}\lambda_{de}^{ac}\right) - [\eta \leftrightarrow f, \Gamma],\qquad(8)$$

$$\frac{d}{ds}\Gamma_{34}^{12} = \sum_a \left(\eta_a^1 \Gamma_{34}^{a2} + \eta_a^2\Gamma_{34}^{1a} - \eta_3^a\Gamma_{a4}^{12} - \eta_4^a\Gamma_{3a}^{12}\right.$$
$$\left. - f_a^1\eta_{34}^{a2} - f_a^2\eta_{34}^{1a} + f_3^a\eta_{a4}^{12} + f_4^a\eta_{3a}^{12}\right)$$
$$+ \frac{1}{2}\sum_{ab}\left(\eta_{ab}^{12}\Gamma_{34}^{ab} - \Gamma_{ab}^{12}\eta_{34}^{ab}\right)(1 - n_a - n_b)$$
$$+ \sum_{ab}(n_a - n_b)\left(\left(\eta_{3b}^{1a}\Gamma_{4a}^{2b} - \Gamma_{3b}^{1a}\eta_{4a}^{2b}\right) - [1 \leftrightarrow 2]\right),\qquad(9)$$

where $E = \langle\Phi| H |\Phi\rangle$, and the one- and two-body parts of H, denoted by f and Γ, contain in-medium contributions from the 3N interaction because of the normal ordering [10, 11]. The symbol $[\eta \leftrightarrow f, \Gamma]$ in Eq. (8) indicates an interchange of the one- and two-body parts of η and H. To close the system of flow equations (7)–(9), we truncate three-body operators [11], as well as a term containing $\lambda^{(3)}$ in the energy flow equation (7). We will refer to this truncation as MR-IM-SRG(2). Note that the two-body flow equations (9) are identical to the closed-shell case [10, 11].

As our default choice for the generator, we use the ansatz of White [11, 15]. The required matrix elements of the Hamiltonian, such as $\langle\Phi| H : A_{34}^{12} : |\Phi\rangle$, which couple the reference state to excitations, or $\langle\Phi| : A_{12}^{34} : H : A_{34}^{12} : |\Phi\rangle$, which enter the energy denominators, can be evaluated using the generalized normal ordering. This yields

$$\eta_2^1 = \frac{\bar{n}_1 n_2 f_2^1}{\bar{n}_1 f_1^1 - n_2 f_2^2 + \bar{n}_1 n_2 \Gamma_{12}^{12}} - [1 \leftrightarrow 2] + \dots,\qquad(10)$$

$$\eta_{34}^{12} = \frac{\bar{n}_1 \bar{n}_2 n_3 n_4 \Gamma_{34}^{12}}{\bar{n}_1 f_1^1 + \bar{n}_2 f_2^2 - n_3 f_3^3 - n_4 f_4^4 + G_{34}^{12}} - [(12) \leftrightarrow (34)]$$
$$+ \dots,\qquad(11)$$

where

$$G_{34}^{12} = \bar{n}_1 \bar{n}_2 \Gamma_{12}^{12} + n_3 n_4 \Gamma_{34}^{34}$$
$$- \left(\bar{n}_1 n_3 \Gamma_{13}^{13} + \bar{n}_2 n_4 \Gamma_{24}^{24} + [1 \leftrightarrow 2]\right).\qquad(12)$$

The dots in Eqs. (10) and (11) indicate terms that are linear in $\lambda^{(2)}$. Terms containing higher powers of $\lambda^{(2)}$ or $\lambda^{(n)}$ with $n \geq 3$ are truncated.

In cases where the flow stalls due to small energy denominators, we use Wegner's generator $\eta = [H, H^{od}]$ as a fallback, defining the one- and two-body parts of the off-diagonal Hamiltonian H^{od} as

$$(f^{od})_2^1 = \bar{n}_1 n_2 f_2^1 + [1 \leftrightarrow 2],$$
$$(\Gamma^{od})_{34}^{12} = \bar{n}_1 \bar{n}_2 n_3 n_4 \Gamma_{34}^{12} + [(12) \leftrightarrow (34)].\qquad(13)$$

This generator is free of numerical instabilities but less efficient because the flow equations become stiff [10, 11]. In the limit of a single Slater determinant reference state, both generators reduce to the forms used for closed-shell nuclei in [10, 11].

As our reference state, we choose a particle-number projected Hartree-Fock-Bogoliubov (HFB) vacuum, $|\Phi\rangle = P_N P_Z |\text{HFB}\rangle$ [16]. This ansatz for the many-body wave function for an open-shell nucleus allows us to enforce spherical symmetry in calculations for even nuclei [17]. The natural-orbital basis of $|\Phi\rangle$ is the usual canonical basis of the HFB vacuum, allowing us to use analytic expressions for the density matrices [18].

We conclude the discussion of the MR-IM-SRG method by mentioning that systematic improvements of the truncation scheme are possible. One would include $3, \dots, A$-body operators when expanding the flow equation (6) in normal-ordered components, as well as additional terms involving irreducible density matrices. While the number of flow equations is the same as in the single-reference case, their complexity grows much more rapidly due to additional terms from the generalized normal ordering [10–12].

Calculation Details. Reference states for the MR-IM-SRG calculation are obtained by solving the HFB equations in 15 major harmonic-oscillator (HO) shells, and projecting the resulting HFB vacuum on good proton and neutron numbers [11, 19]. For the 3N interaction, the sum of the HO energy quantum numbers of a 3N basis state is limited by $e_1 + e_2 + e_3 \leq E_{3\text{max}} = 14$, as discussed in [11, 20]. Reducing $E_{3\text{max}}$ from 14 to 12 changes the MR-IM-SRG(2) ground-state energies for oxygen isotopes by less than 1% for the
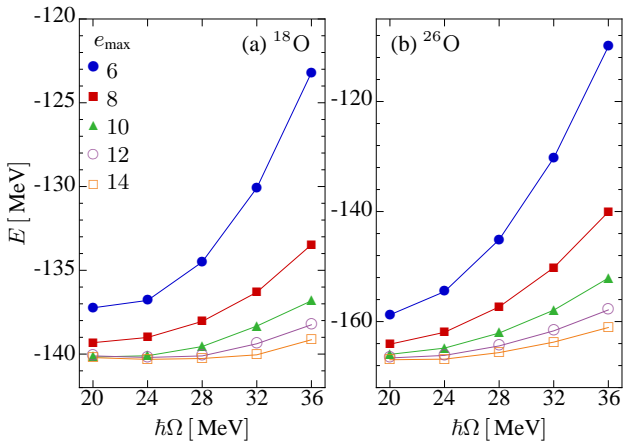
FIG. 1. (Color online) Convergence of the MR-IM-SRG(2) ground-state energies of ^{18}O and ^{26}O with respect to the single-particle basis size $e_{\max}$, for the NN+3N-full Hamiltonian at $\lambda_{\mathrm{SRG}} = 2.0$ fm^{-1}.
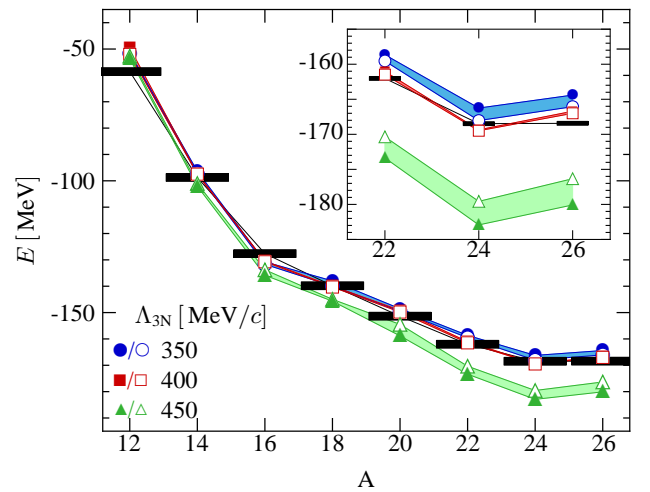

FIG. 2. (Color online) Dependence of the MR-IM-SRG(2) oxygen ground-state energies for the NN+3N-full Hamiltonian on the resolution scale and the initial cutoff $\Lambda_{3\mathrm{N}}$. For each $\Lambda_{3\mathrm{N}}$, the band is obtained by varying λ_{SRG} from 2.24 (open symbols) to 1.88 fm^{-1} (closed symbols). Experimental values are indicated by black bars [24, 25].

Hamiltonians used in this work. The intrinsic NN+3N Hamiltonian is normal-ordered with respect to the reference state, and the residual normal-ordered 3N interaction term is discarded, leading to the normal-ordered two-body approximation (NO2B), which is found to overestimate oxygen binding energies by about 1% [11, 20].

In this Letter, we use the same nuclear Hamiltonians as in our recent IM-SRG and CC studies [11, 20, 21]: The NN interaction is the chiral N^{3}LO interaction by Entem and Machleidt, with cutoff $\Lambda_{\mathrm{NN}} = 500$ MeV/c [2, 22]. Our standard three-body Hamiltonian is a local N^{2}LO 3N interaction with initial cutoff $\Lambda_{3\mathrm{N}} = 400$ MeV/c. The resolution scale of the Hamiltonian is lowered to $\lambda_{\mathrm{SRG}} = 1.88, \ldots, 2.24$ fm^{-1} by means of an SRG evolution in three-body space [23]. Hamiltonians which only contain SRG-induced 3N forces are referred to as NN+3N-induced, those also containing an initial 3N interaction as NN+3N-full.

In Fig. 1, we illustrate the convergence of the MR-IM-SRG(2) ground-state energies for ^{18}O and ^{26}O with respect to the single-particle basis size. At the optimal $\hbar\Omega$, the change in the ground-state energy is of the order of 0.1% when we increase the basis from $e_{\max} = 12$ to 14. This rapid convergence is representative for all Hamiltonians used in this work.

Results. In Fig. 2, we show MR-IM-SRG(2) ground-state energies of the even oxygen isotopes for NN+3N-full Hamiltonians with initial cutoffs $\Lambda_{3\mathrm{N}} = 350, 400$ and 450 MeV/c. For the 3N low-energy constants, we use a fixed $c_D = -0.2$, and $c_E = 0.205, 0.098$, and -0.016, respectively, which are fit to the ^{4}He binding energy in NCSM calculations [21, 26]. For the NN+3N-full Hamiltonian with $\Lambda_{3\mathrm{N}} = 400$ MeV/c, we achieve an excellent reproduction of experimental data all the way to the neutron drip line at ^{24}O [25], with deviations of 1-2%. A recent experiment places the ^{26}O ground-state resonance at $E_x \lesssim 150$ keV above the ^{24}O ground-state energy [27]. We slightly overestimate this energy in our calculation because the HO basis expansion of our single-particle wave functions is ill-suited to the description of resonances

and other continuum states. The inset in Fig. 2 illustrates that the correct drip-line systematics is independent of λ_{SRG} in the studied range. The drip line is also robust against variations of the cutoff $\Lambda_{3\mathrm{N}}$. This suggests that the long-range part of the two-pion exchange (2PE) 3N interaction, which remains unchanged as we lower $\Lambda_{3\mathrm{N}}$, is key to obtaining the proper isotopic trends. The 2PE contribution has significant spin-orbit and tensor terms, and is therefore important for the evolution of the shell structure along the isotopic chain, as also demonstrated in other studies, e.g. [28].

Let us now discuss the effect of varying the resolution scale. As discussed in [11, 20], the λ_{SRG}-dependence of our energies is the net result of omitted induced 4N interactions, the $E_{3\max}$ cut, and the MR-IM-SRG(2) truncation of the many-body expansion, while the effect of the NO2B approximation is found to be independent of λ_{SRG}.

For $\Lambda_{3\mathrm{N}} = 350$ MeV/c we do not expect significant induced 4N interactions [26]. As λ_{SRG} is reduced, we capture additional repulsive 3N strength in matrix elements with $e_1 + e_2 + e_3 \leq E_{3\max}$. We also speed up the convergence of the many-body expansion and reduce the error due to the MR-IM-SRG(2) truncation, but for the resolution scales considered here, this effect is already saturated. In total, we find a slight artificial increase of the ground-state energies as we lower λ_{SRG} [11].

For our standard choice $\Lambda_{3\mathrm{N}} = 400$ MeV/c, effects from omitted 4N interactions, the $E_{3\max}$ cut, and the many-body truncation cancel, and the λ_{SRG}-dependence of the energies in Fig. 2 is extremely weak [11]. The omission of 4N interactions becomes the dominant source of uncertainty as we increase $\Lambda_{3\mathrm{N}}$ to 450 MeV/c, resulting in an enhanced λ_{SRG}-dependence of the ground-state energies of the sd-shell oxygen isotopes. This observation is consistent with the
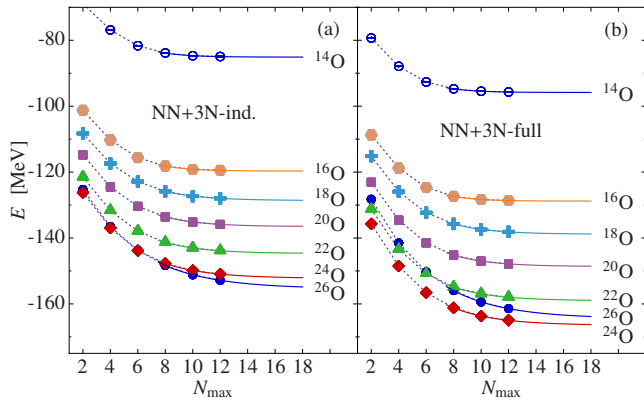
FIG. 3. (Color online) Extrapolation of IT-NCSM ground-state energies for the even oxygen isotopes, obtained using the NN+3N-induced (left) and NN+3N-full Hamiltonians at $\lambda_{\text{SRG}} = 1.88$ fm^{-1}. The solid lines indicate the energy extrapolation based on $N_{\text{max}} = 8 - 12$ data, while dotted lines guide the eye for smaller N_{max}. Uncertainties due to the importance truncation are smaller than the symbols used to represent the data. All energies are obtained at optimal $\hbar\Omega$.

even stronger λ_{SRG}-dependence for $\Lambda_{3N} = 500$ MeV/c, and led us to consider the modified 3N interaction with $\Lambda_{3N} = 400$ MeV/c in the first place [21, 26, 29].

To assess the quality of our MR-IM-SRG(2) ground-state energies, we compare them to results from the IT-NCSM, which yields the exact NCSM results within quantified uncertainties from the importance truncation [30]. In the IT-NCSM calculations, we use the full 3N interaction without NO2B approximation, and the $E_{3\text{max}}$ cut is naturally compatible with the IT-NCSM model space truncation [11]. In Fig. 3 we show the convergence of the oxygen ground-state energies for the NN+3N-induced and NN+3N-full Hamiltonians as a function of N_{max}, along with exponential fits which extrapolate $N_{\text{max}} \to \infty$ [29–31]. With the exception of ^{26}O, all isotopes converge well, and the uncertainties of the threshold and model spaces truncations of the IT-NCSM results are typically of the order of 1 MeV. For ^{26}O, the rate of convergence is significantly worse, which is expected due to the resonance nature of this ground state.

In Fig. 4, we compare the MR-IM-SRG(2) and IT-NCSM ground-state energies of the oxygen isotopes, for the NN+3N-induced and NN+3N-full Hamiltonians with $\lambda_{\text{SRG}} = 1.88$ fm^{-1} to experiment. For the latter, the overall agreement between the two very different many-body approaches and experiment is striking: Except for slightly larger deviations in the drip line nuclei ^{12}O and ^{26}O, we reproduce experimental binding energies within 2-3 MeV. This is a remarkable demonstration of the predictive power of current chiral NN+3N Hamiltonians, at least for ground-state energies. For further confirmation, we perform CC calculations with singles and doubles (CCSD), as well as perturbative triples (Λ-CCSD(T)) [13, 20, 32, 33] for oxygen isotopes with sub-shell closures. Using the same Hamiltonians in NO2B approximation, the MR-IM-SRG energies are bracketed by the CC re-
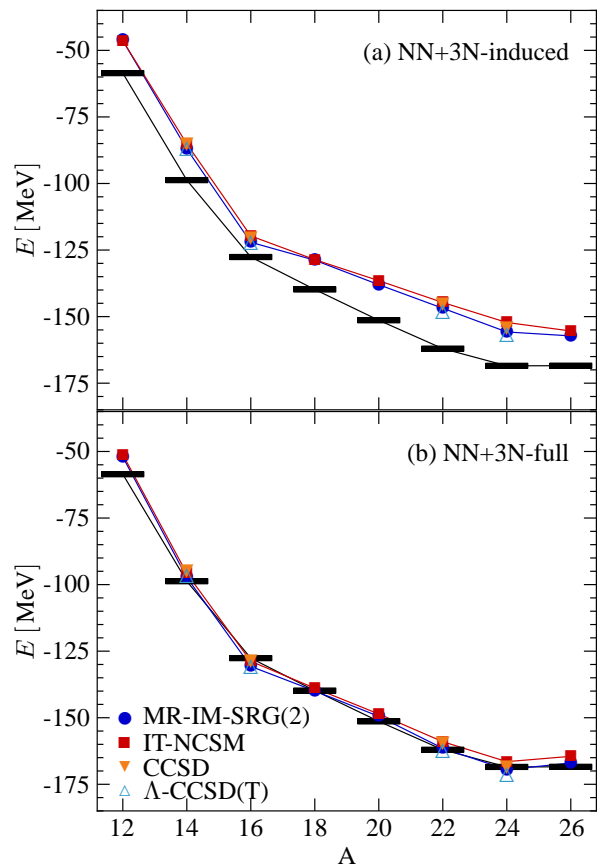

FIG. 4. (Color online) Ground-state energies of the oxygen isotopes from the indicated many-body methods, for the NN+3N-induced (top) and NN+3N-full (bottom) Hamiltonian with $\Lambda_{3N} = 400$ MeV/c. MR-IM-SRG(2), CCSD, and Λ-CCSD(T) results are obtained at optimal $\hbar\Omega$, using 15 major oscillator shells and $E_{3\text{max}} = 14$. The IT-NCSM energies are extrapolations to infinite model space. Experimental values are indicated by black bars [24, 25].

sults, and more similar to the Λ-CCSD(T) values, consistent with the closed-shell results discussed in [11].

For the NN+3N-induced calculation, which should be compared to calculations with the bare chiral NN interaction [6], the reproduction of experimental trends fails, and the neutron drip line is predicted at wrong mass number, because ^{26}O is bound with respect to ^{24}O. This illustrates the crucial importance of the chiral 3N interaction for a proper description of the structure of neutron-rich nuclei [28].

Let us conclude the discussion by addressing the uncertainties of our results. The MR-IM-SRG(2) energies lie about 1.5–2% below the IT-NCSM results. About 1% of this deviation is caused by the NO2B approximation. The uncertainty due to the $E_{3\text{max}}$ cut should be smaller than 1% at low λ_{SRG}. While the NO2B and $E_{3\text{max}}$ uncertainties exhaust the greater part of the 1.5–2% deviation between MR-IM-SRG(2) and IT-NCSM, and suggest a very small uncertainty due to the many-body truncation, we assume a more conservative many-body truncation error of 1–1.5%, and an overall uncertainty of our oxygen energies at the level of 3–3.5%, consistent with

our closed-shell IM-SRG calculations [11]. Because all irreducible many-body density matrices vanish in closed-shell nuclei, our findings indicate that the truncation of terms containing $\lambda^{(3)}$ and higher, or non-linear powers of $\lambda^{(2)}$, is negligible compared to the truncation of induced three-body operators. A more detailed analysis of the MR-IM-SRG truncation scheme will be presented in a future publication.

Conclusions. We have generalized the IM-SRG approach to multi-reference states, and used the resulting MR-IM-SRG method to perform the first *ab initio* study of the even oxygen isotopes with chiral NN+3N Hamiltonians, along with the IT-NCSM and the CC method. We achieve remarkable agreement with experimental binding energies within theoretical uncertainties of 3%. This agreement is achieved without any re-adjustment of the interaction to experimental data beyond ^{4}He, and therefore constitutes an impressive demonstration of the predictive power of chiral NN+3N Hamiltonians. The present work also highlights the importance of the 3N interaction for the nuclear structure of neutron-rich nuclei, as demonstrated by the robust reproduction of the oxygen drip line.

While the oxygen isotopes studied here are also within the reach of the particle-attached and -removed CC calculations, the MR-IM-SRG provides ground-state energies at a significantly reduced computational cost. Thus, it is ideally suited for calculations in medium- and eventually heavy-mass open-shell nuclei far from shell closures. In such applications, the MR-IM-SRG also offers a more complete treatment of many-body correlations than other current open-shell methods.

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