Ab-Initio Coupled-Cluster Study of ^{16}O

M. Włoch,[1] D. J. Dean,[2] J. R. Gour,[1] M. Hjorth-Jensen,[3] K. Kowalski,[1] T. Papenbrock,[2,4] and P. Piecuch[1]

[1]*Department of Chemistry, Michigan State University, East Lansing, Michigan 48824, USA*
[2]*Physics Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831, USA*
[3]*Department of Physics and Center of Mathematics for Applications, University of Oslo, N-0316 Oslo, Norway*
[4]*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA*

(Received 26 January 2005; published 3 June 2005)

We report converged results for the ground and excited states and matter density of ^{16}O using realistic two-body nucleon-nucleon interactions and coupled-cluster methods and algorithms developed in quantum chemistry. Most of the binding is obtained with the coupled-cluster singles and doubles approach. Additional binding due to three-body clusters (triples) is minimal. The coupled-cluster method with singles and doubles provides a good description of the matter density, charge radius, charge form factor, and excited states of a one-particle, one-hole nature, but it cannot describe the first-excited 0^+ state. Incorporation of triples has no effect on the latter finding.

DOI: 10.1103/PhysRevLett.94.212501 PACS numbers: 21.60.−n, 21.10.−k, 27.20.+n

One of the most important problems in nuclear physics is to understand how nuclear properties arise from the underlying nucleon-nucleon interactions. Recent progress using Monte Carlo [1] and diagonalization [2] techniques produced converged results for nuclei with up to $A = 12$ active particles, yielding a much-improved understanding of nuclear forces in light systems. To extend these *ab-initio* studies to medium-size nuclei, one must explore computationally less expensive methods. Coupled-cluster theory [3] (see [4–7] for reviews) is a promising candidate for such developments since it can provide a highly accurate description of many-particle correlations at relatively low cost. Recently, Mihaila and Heisenberg performed coupled-cluster calculations for the binding energy and the electron scattering form factor of ^{16}O using modern interactions and bare Hamiltonian [8] (for a review of earlier applications of coupled-cluster theory to nuclei, see, e.g., [4]). In previous work [9], we took another route and used quantum chemical coupled-cluster methods and the renormalized Hamiltonian to compute ground and excited states of ^{4}He and ground-state energies of ^{16}O in a small model space consisting of 4 major oscillator shells, demonstrating promising results when compared with exact shell-model diagonalization.

In this Letter we report, for the first time, converged coupled-cluster calculations for ground-state and excited-state energies and other properties of ^{16}O using modern nucleon-nucleon interactions derived from effective-field theory [10]. Our ground-state calculations involving one-body and two-body components of the cluster operator are performed in up to 8 major oscillator shells (480 uncoupled single-particle basis states), while the corrections due to three-body clusters and computations of excited states and nuclear properties involve up to 7 major oscillator shells (336 single-particle states). The significant progress in going from model calculations using 80 single-particle states [9] to large-scale calculations involving 16 correlated nucleons and almost 500 single-particle states has been possible due to the development of general-purpose coupled-cluster computer programs for nuclear structure, using diagram factorization techniques and algorithms to solve coupled-cluster equations from quantum chemistry. We pay particular attention to three aspects of the calculations: (i) the convergence of the ground-state energy with respect to the size of the model space and the role of higher−than−two-body clusters in such studies, (ii) the ability of coupled-cluster methods to describe excited states, and (iii) the performance of coupled-cluster methods in studies of nuclear radii, matter density, charge form factor, and occupation numbers. We have not yet included the three-nucleon interaction that should eventually be considered [1,2]. However, our calculations represent a dramatic step forward in nuclear many-body computations due to the enormous oscillator space we probe through application of computationally efficient coupled-cluster methods. They teach us about the nucleon correlations and the magnitude of the (missing) three-body forces.

We use two variants of effective-field-theory-inspired Hamiltonians, Idaho-A and N^{3}LO [11]. The Idaho-A potential was derived with up to chiral-order three diagrams while N^{3}LO includes chiral-order four diagrams, and charge-symmetry and charge-independence breaking terms. We also include the Coulomb interaction with the N^{3}LO calculations. Since very slow convergence with the number of single-particle basis states was obtained using bare interactions [8], we renormalize the bare Hamiltonian using a no-core G-matrix approach [12] which obtains a starting-energy dependence $\tilde{\omega}$ in the two-body matrix elements $G(\tilde{\omega})$. We use the Bethe-Brandow-Petschek [13] theorem to alleviate much of the starting-energy dependence (see [12] for details). This dependence is weak for ^{16}O, particularly for the matrix elements below the Fermi surface [14]. The effective Hamiltonian for coupled-cluster calculations is $H' = t + G(\tilde{\omega})$, where t is the kinetic energy. We correct H' for center-of-mass contaminations using the expression $H = H' + \beta_{\rm c.m.} H_{\rm c.m.}$. We choose $\beta_{\rm c.m.}$ such that the expectation value of the center-of-mass Hamiltonian $H_{\rm c.m.}$ is 0.0 MeV. We note that intrinsic excitation energies are virtually independent of $\beta_{\rm c.m.}$ while the unphysical, center-of-mass contaminated states show a sharp, nearly linear dependence of excitation energies on $\beta_{\rm c.m.}$. This allows us to separate intrinsic and center-of-mass contaminated states.

Once the one- and two-body matrix elements of the Hamiltonian H are constructed, we solve the A-body problem using quantum chemical coupled-cluster techniques. In the ground-state calculations, we use the coupled-cluster singles and doubles (CCSD) approach [15], to describe correlation effects due to one- and two-body clusters, and the completely renormalized CCSD(T) [CR-CCSD(T)] method [16], to correct the CCSD energies for the effects of three-body clusters (''triples''). In the excited-state and property calculations, we use the equation-of-motion (EOM) CCSD method [17] (equivalent to the linear response CCSD approach [18]). We also correct the energies of excited states obtained with EOMCCSD for the effects of triples using the CR-EOMCCSD(T) approach [16]. The details of the above methods can be found elsewhere [15–17]. Here, we only mention that the CCSD method is obtained by truncating the many-body expansion for the cluster operator T in the exponential ansatz exploited in coupled-cluster theory, $|\Psi_0\rangle = \exp(T)|\Phi\rangle$, where $|\Psi_0\rangle$ is the correlated ground-state wave function and $|\Phi\rangle$ is the reference determinant. Following Ref. [9], we take the oscillator product state of ^{16}O as the reference state. The truncated cluster operator used in the CCSD calculations has the form $T = T_1 + T_2$, where $T_1 = \sum_{i,a} t_a^i a_a^\dagger a_i$, and $T_2 = \frac{1}{4}\sum_{ij,ab} t_{ab}^{ij} a_a^\dagger a_b^\dagger a_j a_i$ are the singly and doubly excited clusters and $i, j, \ldots$ $(a, b, \ldots)$ label the single-particle states occupied (unoccupied) in $|\Phi\rangle$. We determine the singly and doubly excited cluster amplitudes t_a^i and t_{ab}^{ij} by solving the nonlinear system of algebraic equations, $\langle\Phi_i^a|\bar{H}|\Phi\rangle = 0$, $\langle\Phi_{ij}^{ab}|\bar{H}|\Phi\rangle = 0$, where $\bar{H} = \exp(-T)H\exp(T)$, and $|\Phi_i^a\rangle$ and $|\Phi_{ij}^{ab}\rangle$ are the singly and doubly excited determinants, respectively, relative to $|\Phi\rangle$. We calculate the ground-state energy E_0 as $\langle\Phi|\bar{H}|\Phi\rangle$. We diagonalize the similarity-transformed Hamiltonian $\bar{H}$ in the relatively small space of singly and doubly excited determinants $|\Phi_i^a\rangle$ and $|\Phi_{ij}^{ab}\rangle$ to obtain the excited-state wave functions $|\Psi_\mu\rangle$ and energies E_μ. The right eigenstates of $\bar{H}$, $R^{(\mu)}|\Phi\rangle$, where $R^{(\mu)} = R_0 + R_1 + R_2$ is a sum of the relevant reference (R_0), one-body (R_1), and two-body (R_2) components define the excited-state ''ket'' wave functions $|\Psi_\mu\rangle = R^{(\mu)}\exp(T)|\Phi\rangle$, whereas the left eigenstates $\langle\Phi|L^{(\mu)}$ define the ''bra'' wave functions $\langle\tilde{\Psi}_\mu| = \langle\Phi|L^{(\mu)}\exp(-T)$. Here, each n-body component of $R^{(\mu)}$ with $n > 0$ is a particle-hole *excitation* operator similar to T_n, whereas $L^{(\mu)}$ is a hole-particle *deexcitation* operator, so that $L_1 = \sum_{i,a} l_i^a a_i^\dagger a_a$ and $L_2 = \frac{1}{4}\sum_{ij,ab} l_{ij}^{ab} a_i^\dagger a_j^\dagger a_b a_a$. The right and left eigenstates of $\bar{H}$ form a biorthonormal set, $\langle\Phi|L^{(\mu)}R^{(\nu)}|\Phi\rangle = \delta_{\mu\nu}$. If the only purpose of the calculation is to obtain excitation energies, the left eigenstates $\langle\Phi|L^{(\mu)}$ are not needed. However, for properties other than energy, both right and left eigenstates of $\bar{H}$ are important. In particular, we calculate the one-body reduced density matrix $\rho_{\alpha\beta}$ in quantum state $|\Psi_\mu\rangle$ as follows:

$$\rho_{\alpha\beta} = \langle\Phi|L^{(\mu)}[\exp(-T)a_\alpha^\dagger a_\beta \exp(T)]R^{(\mu)}|\Phi\rangle. \quad (1)$$

In the CCSD ground-state ($\mu = 0$) case, we have $T = T_1 + T_2$, $R^{(0)} = 1$, and $L^{(0)} = 1 + \Lambda_1 + \Lambda_2$, where the one-body and two-body deexcitation operators Λ_1 and Λ_2 are determined by solving the CCSD left eigenvalue problem, obtained by right-projecting the equation $\langle\Phi|(1+\Lambda)\bar{H} = E_0\langle\Phi|(1+\Lambda)$, with E_0 representing the CCSD energy and $\Lambda = \Lambda_1 + \Lambda_2$, on the singly and doubly excited determinants. Thus far, we have focused on the CCSD and EOMCCSD methods which use inexpensive computational steps that scale as $n_o^2 n_u^4$, where n_o (n_u) is the number of occupied (unoccupied) single-particle states. While the full inclusion of triply excited clusters is possible, the resulting methods are expensive and scale as $n_o^3 n_u^5$. Thus, we estimate the effects of T_3 and R_3 on ground-state and excited-state energies by adding the corrections to the CCSD or EOMCCSD energies, which only require $n_o^3 n_u^4$ noniterative steps. These corrections, due to T_3 and R_3, define the CR-CCSD(T) and CR-EOMCCSD(T) approaches [7,16]. In this study, we use variant ''c'' of the corresponding approaches, as described in [9].

We turn to a discussion of our ^{16}O results. We choose the oscillator energy $\hbar\omega$ for our basis states to minimize the CCSD energy. For the $N = 7$ and $N = 8$ oscillator shell runs, $\hbar\omega = 11$ MeV, and the results are nearly independent of $\hbar\omega$ [12]. Shown in Fig. 1 are our CCSD or EOMCCSD and CR-CCSD(T) or CR-EOMCCSD(T) ground-state and excited-state energies as a function of N. The symbols in Fig. 1 represent our calculations while

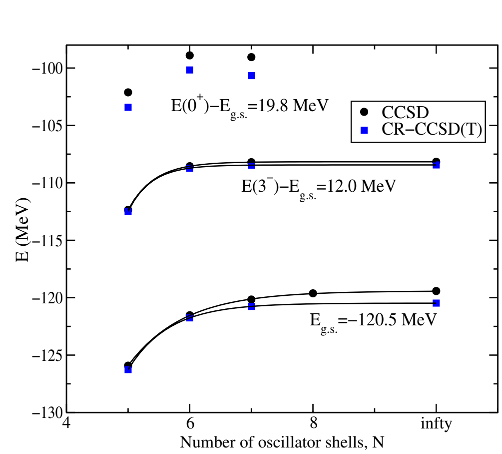

FIG. 1 (color online). The coupled-cluster energies of the ground-state (g.s.) and first-excited 3^- and 0^+ states as functions of the number of oscillator shells N obtained with the Idaho-A interaction.

the lines represent a fit of the form $E(N) = E_\infty + a\exp(-bN)$, where the extrapolated energy E_∞ and a and b are parameters for the fit. We also show in Fig. 1 our calculations for the first-excited 3^- state and the position of the lowest calculated 0^+ excited state. We now discuss these results.

Triples correction to the CCSD ground-state energy.—The small model space calculation [9] indicates that the triples corrections to the ground-state CCSD energies are small. We extend these calculations from 4 to 8 major oscillator shells for CCSD calculations and to 7 major oscillator shells for CR-CCSD(T) calculations, as shown in Fig. 1. We find that the extrapolated CCSD energy is -119.4 MeV for Idaho-A. For the $N = 7$ Idaho-A calculation, the difference between the CCSD and CR-CCSD(T) result is 0.6 MeV, while the extrapolated values differ by only 1.1 MeV; our extrapolated CR-CCSD(T) energy is -120.5 MeV. The Coulomb interaction adds to the binding 11.2 MeV, so that our estimated Idaho-A ground-state energy is -109.3 MeV (compared to an experimental value of -128 MeV). Our $N = 7$ ($N = 8$) N^3LO CCSD and CR-CCSD(T) energies, which include the Coulomb interaction, are -112.4 (-111.2) and -112.8 (-112.0) MeV, respectively. Thus, the two-body interactions underbind ^{16}O by approximately 1 MeV per particle, pointing to the need for three-body forces. For the Idaho-A and N^3LO interactions and the ^{16}O nucleus, we conclude that connected T_3 clusters are indeed small, contributing less than 1% to the ground-state energy. This is an important finding, since it implies that ground-state correlations of a closed-shell nucleus with two-nucleon interactions can be captured by the relatively inexpensive CCSD approach. Another important finding is a rapid convergence of the CCSD and CR-CCSD(T) energies with the number of oscillator shells due to the renormalized form of the Hamiltonian. For example, the difference between the $N = 8$ and $N = 7$ CCSD or Idaho-A energies is 0.5 MeV (see Fig. 1).

Calculations of the first-excited 3^- *state.*—The first-excited 3^- state in ^{16}O is thought to be principally a one-particle–one-hole ($1p$-$1h$) state [19]. The experience of quantum chemistry is that the EOMCCSD and CR-EOMCCSD(T) methods describe such states well (provided that three-body interactions can be ignored). The largest R_1 amplitudes obtained in the EOMCCSD calculations indicate that the dominant $1p$-$1h$ excitations are from the $0p_{1/2}$ orbital to the $0d_{5/2}$ orbital. The $2p$-$2h$ excitations in the EOMCCSD wave function, defined as $R_2 + R_1T_1 + R_0(T_2 + T_1^2/2)$ ($R_0 = 0$ in this case), are much smaller than the R_1 amplitudes, and the CR-EOMCCSD(T) calculation hardly changes the total energy of the state, which indicates that this state has indeed a $1p$-$1h$ nature. Our extrapolated Idaho-A results indicate that the 3^- state lies at -108.2 and -108.4 MeV in the EOMCCSD and CR-EOMCCSD(T) calculations, respectively. The CR-EOMCCSD(T) method yields an excitation energy of 12.0 MeV for this state which experimentally lies at 6.13 MeV. N^3LO yields similar results. Based on the $1p$-$1h$ structure of the state, we conclude that Idaho-A and N^3LO do not yield an excitation energy for the 3^- state which is commensurate with experiment. These results agree with recent no-core shell-model calculations with similar two-body Hamiltonians [20]. The 3^- state is expected to be built on $1p$-$1h$ excitations which depend on the single-particle splittings. These splittings will be affected by three-body forces not included in our Hamiltonian, thus affecting the energy of the 3^- state. Whether other mechanisms than three-body forces can provide an additional binding of 6 MeV needs further research. Our results are converged at the coupled-cluster level employing the Idaho-A and N^3LO two-body interactions, so it is likely that the discrepancy between theory and experiment resides in the Hamiltonian, not in the correlation effects which EOMCCSD and CR-EOMCCSD(T) describe very well if three-body forces play no role and if the state has a $1p$-$1h$ nature.

Calculation of the first-excited 0^+ *state.*—This state (experimentally at 6.05 MeV), believed to have a $4p$-$4h$ character, cannot be described by our methods. This is confirmed by the large differences between the EOMCCSD or CR-EOMCCSD(T) results and experiment (see Fig. 1). One would need to include $4p$-$4h$ cluster operators (T_4 and R_4) to improve our results.

We also performed preliminary calculations for other negative parity states. The quartet of negative parity states starting with the $J = 3^-$ state, and including the $J = 1^-$, 2^-, and 0^- states, are all believed to have a similar $1p$-$1h$ character [19]. The EOMCCSD calculation with 5 major oscillator shells and Idaho-A confirms the existence of this quartet, giving excitation energies of 13.57, 15.37, 17.07, and 17.15 MeV for the $J = 3^-$, 1^-, 2^-, and 0^- states, respectively. While these states are all a few MeV above the experimental values, their ordering predicted by EOMCCSD is correct.

Calculation of the one-body density.—We use Eq. (1), where $\mu = 0$, to calculate the radial ground-state density $\rho(r)$ and the root-mean-square (rms) radius of ^{16}O (Fig. 2). After correcting for the finite sizes of the nucleons, which experimentally are $r_p^2 = 0.743$ fm^2 and $r_n^2 = 0.115$ fm^2, and for the $0s$ center-of-mass motion, for which we use $\langle \Psi_0 \mid \mathbf{R} \mid \Psi_0 \rangle = \frac{62.2071}{A\hbar\omega}$ fm^2, our rms charge radii for ^{16}O for 5, 6, and 7 oscillator shells are 2.45, 2.50, and 2.51 fm, respectively, when the Idaho-A potential is used (N^3LO gives similar values). The experimental charge radius is 2.73 ± 0.025 fm. We also calculate the occupation probability for the natural orbitals. Experimental data from quasielastic proton knockout [21] yields 2.17 ± 0.12% for the $0d_{5/2}$ occupation and 1.78 ± 0.36% for the $1s_{1/2}$ occupation. We obtain 3.2% and 2.3%, respectively, using Idaho-A in the $N = 7$ model space. For N^3LO in the $N = 7$ model space, we obtain 3.8% and 2.6%, respectively. For the calculation of the nuclear charge form factor, we follow

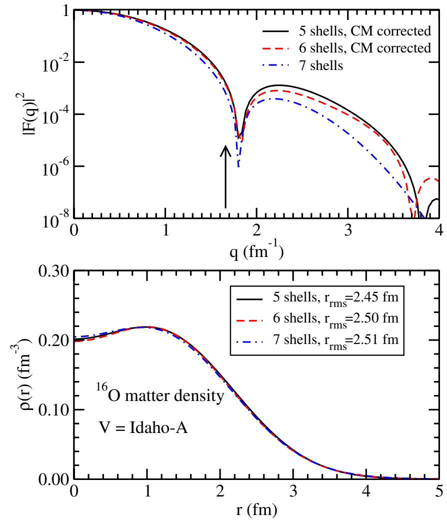

FIG. 2 (color online). Top panel: The charge form factor computed from the CCSD density matrix. Bottom panel: the matter density in ^{16}O. The results obtained with the Idaho-A interaction.

[22]. In this approach, the form factor includes contributions from the two-body reduced density matrix due to center-of-mass corrections. We compute the one-body density contributions within the framework of CCSD theory using Eq. (1). The contributions of the two-body density matrix are computed within the shell-model like description as $\rho_{\alpha\beta\gamma\delta} = \langle\Psi_0|\, a_\alpha^\dagger a_\beta^\dagger a_\delta a_\gamma \,|\Psi_0\rangle / \langle\Psi_0|\Psi_0\rangle$, where we approximate $|\Psi_0\rangle$ by $(1 + C_1 + C_2)\,|\,\Phi\rangle$, with $C_1 = T_1$ and $C_2 = T_2 + \frac{1}{2}T_1^2$ defining the $1p-1h$ and $2p-2h$ components of the CCSD wave function. The upper part of Fig. 2 shows the charge form factor for different model spaces. The 5-shell and 6-shell results include the center-of-mass corrections and exhibit a second zero. Compared to the experimental value (the arrow in Fig. 2), the first zero of the form factor is reasonable, although slightly too large; this is consistent with an underestimated value of the theoretical charge radius.

In summary, the ^{16}O ground state is converged with respect to the model space size and is accurately described within the basic CCSD approximation, with three-body clusters contributing less than 1% of the binding energy. We attribute the 1 MeV per particle difference between the coupled-cluster and experimental binding energies to three-body forces. We obtained a correct description of the quartet of low-lying negative parity $1p$-$1h$ excited states, although there is a 6 MeV difference between the converged coupled-cluster results and experiment for the lowest $J = 3^-$ state, which is, quite likely, due to an inadequate description of the relevant nuclear forces by the Hamiltonian. We were unable to accurately describe the lowest $J = 0^+$ excited state due to connected $4p$-$4h$ correlations missing in coupled-cluster approximations employed in this study. The CCSD method provides reasonable results for the nuclear matter density, charge radius, and charge form factor. The use of the renormalized Hamiltonian guarantees fast convergence of the results with the number of oscillator shells. All of this makes low-cost coupled-cluster methods a promising alternative to traditional shell-model diagonalization techniques.

Research supported by the U.S. Department of Energy (Oak Ridge National Laboratory, University of Tennessee, Michigan State University), the National Science Foundation (Michigan State University), and the Research Council of Norway (University of Oslo).

[1] R. B. Wiringa and S. C. Pieper, Phys. Rev. Lett. **89**, 182501 (2002).
[2] P. Navrátil and W. E. Ormand, Phys. Rev. C **68**, 034305 (2003).
[3] F. Coester, Nucl. Phys. **7**, 421 (1958); F. Coester and H. Kümmel, Nucl. Phys. **17**, 477 (1960); J. Čížek, J. Chem. Phys. **45**, 4256 (1966); Adv. Chem. Phys. **14**, 35 (1969).
[4] H. Kümmel, K. H. Lührmann, and J. G. Zabolitzky, Phys. Rep. **36**, 1 (1978).
[5] H. G. Kümmel, in *Recent Progress in Many-Body Theories*, edited by R. F. Bishop *et al.*, (World Scientific, Singapore, 2002), p. 334; R. F. Bishop, in *Microscopic Quantum Many-Body Theories and Their Applications*, edited by J. Navarro and A. Polls, Lecture Notes in Physics, Vol. 510 (Springer, Berlin, 1998), p. 1.
[6] R. J. Bartlett, J. Phys. Chem. **93**, 1697 (1989); J. Paldus and X. Li, Adv. Chem. Phys. **110**, 1 (1999); T. D. Crawford and H. F. Schaefer III, Rev. Comput. Chem. **14**, 33 (2000).
[7] P. Piecuch *et al.*, Theor. Chem. Acc. **112**, 349 (2004).
[8] J. H. Heisenberg and B. Mihaila, Phys. Rev. C **59**, 1440 (1999); B. Mihaila and J. H. Heisenberg, Phys. Rev. C **61**, 054309 (2000).
[9] K. Kowalski *et al.*, Phys. Rev. Lett. **92**, 132501 (2004).
[10] S. Weinberg, Phys. Lett. B **251**, 288 (1990); U. van Kolck, Prog. Part. Nucl. Phys. **43**, 337 (1999).
[11] D. R. Entem and R. Machleidt, Phys. Lett. B **524**, 93 (2002);Phys. Rev. C **68**, 41001 (2003).
[12] D. J. Dean and M. Hjorth-Jensen, Phys. Rev. C **69**, 054320 (2004).
[13] H. A. Bethe, B. H. Brandow, and A. G. Petschek, Phys. Rev. **129**, 225 (1963).
[14] P. J. Ellis *et al.*, Nucl. Phys. **A573**, 216 (1994).
[15] G. D. Purvis and R. J. Bartlett, J. Chem. Phys. **76**, 1910 (1982).
[16] K. Kowalski and P. Piecuch, J. Chem. Phys. **120**, 1715 (2004).
[17] J. F. Stanton and R. J. Bartlett, J. Chem. Phys. **98**, 7029 (1993); P. Piecuch and R. J. Bartlett, Adv. Quantum Chem. **34**, 295 (1999).
[18] H. Monkhorst, Int. J. Quantum Chem., Quantum Chem. Symp. **11**, 421 (1977); K. Emrich, Nucl. Phys. **A351**, 379 (1981).
[19] E. K. Warburton and B. A. Brown, Phys. Rev. C **46**, 923 (1992).
[20] P. Navrátil and J. P. Vary, (private communication).
[21] M. Leuschner *et al.*, Phys. Rev. C **49**, 955 (1994).
[22] B. Mihaila and J. H. Heisenberg, Phys. Rev. Lett. **84**, 1403 (2000); Phys. Rev. C **60**, 054303 (1999).