NATURAL ORBITALS FOR THE EQUATION OF MOTION PHONON METHOD*

P. Veselý

Nuclear Physics Institute, Czech Academy of Sciences, Řež, Czechia

G. De Gregorio

INFN Sezione di Napoli, Naples, Italy
and
Dipartimento di Matematica e Fisica
Università degli Studi della Campania Luigi Vanvitelli, 81100 Caserta, Italy

F. Knapp

Faculty of Mathematics and Physics, Charles University, Prague, Czechia

N. Lo Iudice

Dipartimento di Fisica, Universita di Napoli "Federico II", Napoli, Italy

J. Pokorný

Faculty of Nuclear Sciences and Physical Engineering
Czech Technical University in Prague, Prague, Czechia

(Received January 7, 2020)

We discuss the use of natural orbitals as single-particle basis states for the Equation of Motion Phonon Method (EMPM). They are obtained by computing a ground-state one-body density matrix in second-order many-body perturbation theory. We focus our attention on energy and proton point radius of ^{16}O and show that, with respect to Hartree–Fock, the new basis improves drastically the convergence of the two-phonon correlation energy.

DOI:10.5506/APhysPolB.51.623

* Presented at the XXXVI Mazurian Lakes Conference on Physics, Piaski, Poland, September 1–7, 2019.

1. Introduction

Practically all *ab initio* studies of nuclear properties require the solution of the many-body eigenvalue problem in spaces of very large dimensions. It is, therefore, crucial to search for recipes, which allow an effective truncation of the many-body space with no detriment to the accuracy of the solution.

A viable route consists in acting at the mean-field level and trying to find a single-particle basis, which absorbs an appreciable part of the correlations induced by the nuclear interaction. Hartree–Fock (HF) provides a variational optimization of the basis states below the Fermi surface. The unoccupied states, however, are just fixed by imposing the orthonormalization to the occupied states. Moreover, when applied to modern potentials, HF underestimates the binding energy.

Recently, it has been proposed [1, 2] to replace the HF with the eigenvectors of the one-body density matrix, known as natural (NAT) orbitals, adopted in quantum chemistry and atomic physics [3]. No-core shell-model (NCSM) calculations, exploiting the formalism of Ref. [4], were performed in spaces with increasing dimensions for different harmonic oscillator (HO) frequencies [2]. It was found that this new basis is much less sensitive to the dependence of the harmonic oscillator frequency and improves the convergence with the space dimensions.

Here, we intend to investigate if an analogous result is achieved when natural orbitals are used in calculations performed within the equation of motion phonon method (EMPM). This approach [6–8] generates a basis of correlated states composed of tensor products of phonons generated in the Tamm–Dancoff approximation (TDA) and uses such a basis to diagonalize a realistic Hamiltonian. It was used to study the bulk properties of closed-shell nuclei [9] as well as the spectroscopy of even [10, 11] and odd [12–14] nuclei.

2. Theoretical framework

Following Ref. [2], we generate a HF basis by adopting an intrinsic Hamiltonian of the form of

$$H = T + V^{NN} - T_{\mathrm{cm}}\,, \tag{1}$$

where T and T_{cm} are the nucleon and center-of-mass (c.m.) kinetic energies, respectively, while V^{NN} is the nucleon–nucleon (NN) two-body potential. We do not include three-body forces, which, instead, were considered in Ref. [2].

The HF basis is used to compute in second-order many-body perturbation theory (MBPT) the ground-state one-body density matrix

$$\rho_{ij} = \left\langle \Psi_0^{(2)} \right| a_i^\dagger a_j \left| \Psi_0^{(2)} \right\rangle \tag{2}$$

under the constraint

$$\mathrm{Tr}\rho = \sum_i \rho_{ii}^{\mathrm{tot}} = A\,. \tag{3}$$

In the above equations, A denotes the total number of nucleons, $|\Psi_0^{(2)}\rangle$ is the ground-state wave function up to second order, and the operators $a_p^\dagger$ (a_p) and a_h ($a_h^\dagger$) create (annihilate), respectively, a particle and a hole of energy ϵ_p and ϵ_h with respect to the HF vacuum.

The resulting one-body density matrix (2) can be decomposed as follows [4]:

$$\rho_{ij} \simeq \rho_{ij}^{00} + \rho_{ij}^{11} + \rho_{ij}^{20} + \rho_{ij}^{02}\,, \tag{4}$$

where ρ^{00} is the zero-order HF density and the higher-order terms are given by

$$\rho_{h_i h_j}^{11} = -\frac{1}{2} \sum_{p_1 p_2 h_2} \frac{H_{p_1 p_2 h_j h_2}}{\left(e_{h_j} + e_{h_2} - e_{p_1} - e_{p_2}\right)} \frac{H_{p_1 p_2 h_i h_2}}{\left(e_{h_i} + e_{h_2} - e_{p_1} - e_{p_2}\right)}\,, \tag{5}$$

$$\rho_{p_i p_j}^{11} = \frac{1}{2} \sum_{h_1 h_2 p_2} \frac{H_{p_i p_2 h_2 h_1}}{\left(e_{h_1} + e_{h_2} - e_{p_i} - e_{p_2}\right)} \frac{H_{p_j p_2 h_2 h_1}}{\left(e_{h_1} + e_{h_2} - e_{p_j} - e_{p_2}\right)}\,, \tag{6}$$

$$\begin{aligned} \rho_{p_i h_j}^{20} = {} & \frac{1}{2} \sum_{p_1 h_1 p_2} \frac{H_{p_i h_1 p_1 p_2}}{\left(e_{h_1} + e_{h_j} - e_{p_1} - e_{p_2}\right)} \frac{H_{p_1 p_2 h_j h_1}}{\left(e_{h_j} - e_{p_i}\right)} \\ & - \frac{1}{2} \sum_{p_1 h_1 h_2} \frac{H_{p_i p_1 h_1 h_2}}{\left(e_{h_1} + e_{h_2} - e_{p_1} - e_{p_i}\right)} \frac{H_{h_1 h_2 h_j p_1}}{\left(e_{h_j} - e_{p_i}\right)}\,. \end{aligned} \tag{7}$$

The terms ρ_{ij}^{10} vanish because of the Brillouin theorem [5]. The terms ρ^{12} and ρ^{22} have been neglected.

The diagonalization of the one-body density matrix (2) yields the basis of natural orbitals. We define the new creation and annihilation operators $b_i^\dagger$ and b_j, which are linear combinations of the corresponding HF operators, as well as the new vacuum

$$|\mathrm{NAT}\rangle = \Pi_h b_h^\dagger \,|\,\rangle\,, \tag{8}$$

where h labels all occupied states. Given the correlated nature of $|\mathrm{NAT}\rangle$, we need to redefine the new hole states as the eigenstates of ρ with the largest occupation number. We are now able to construct NAT particle–hole states and generate NAT TDA phonons

$$O_\lambda^\dagger = \sum_{ph} c_{ph}^\lambda \left(b_p^\dagger \times b_h\right)^\lambda \tag{9}$$

of energy E_λ. These phonons are the constituents of the EMPM states.

In fact, the primary goal of the EMPM is to generate a basis of n-phonon correlated states of the form of

$$|\alpha_n\rangle = \sum_{\lambda\alpha_{n-1}} C^{\alpha_n}_{\lambda\alpha_{n-1}} \, |(\lambda \times \alpha_{n-1})^{\alpha_n}\rangle = \sum_{\lambda\alpha_{n-1}} \left| \left(O^\dagger_\lambda \times \alpha_{n-1} \right)^{\alpha_n} \right\rangle , \qquad (10)$$

where the TDA phonon operators $O^\dagger_\lambda$ act on the $(n-1)$-phonon basis states $|\alpha_{n-1}\rangle$ of energy $E_{\alpha_{n-1}}$, assumed to be known.

In order to determine $|\,\alpha_n\rangle$, we start [8] with the equations of motion

$$\langle\beta|| \left[H, O^\dagger_\lambda \right]^\lambda ||\alpha\rangle = (E_\beta - E_\alpha) \langle\beta|| \, O^\dagger_\lambda \, ||\alpha\rangle \, , \qquad (11)$$

where we have put $\beta = \alpha_n$ and $\alpha = \alpha_{n-1}$ for simplicity. After expanding the commutator and performing several manipulations, we obtain the generalized eigenvalue equation

$$\sum_{\lambda'\alpha'\lambda''\alpha''} \left[(E_\lambda + E_\alpha - E_\beta)\delta_{\lambda\lambda'}\delta_{\alpha\alpha'} + \mathcal{V}^\beta_{\lambda\alpha\lambda'\alpha'} \right] \mathcal{D}^\beta_{\lambda'\alpha'\lambda''\alpha''} C^\beta_{\lambda''\alpha''} = 0 \, , \qquad (12)$$

where $\mathcal{V}^\beta_{\lambda\alpha\lambda'\alpha'}$ is a phonon–phonon potential [8], and $\mathcal{D}^\beta_{\lambda\alpha\lambda'\alpha} = \langle(\lambda\times\alpha)^\beta|(\lambda'\times\alpha')^\beta\rangle$ is the overlap matrix, which re-introduces the exchange terms among different phonons, re-establishing thereby the Pauli principle.

Equation (12) is ill-defined. In fact, the overlap matrix $\mathcal{D}$ is singular, since the set of states $|(\lambda \times \alpha)^\beta\rangle$ is overcomplete. We adopt the Cholesky decomposition method to extract a basis of linearly-independent states spanning the subspace of the correct dimensions [6, 7]. Thus, the generalized eigenvalue equation can be solved and yields the eigenstates $|\alpha_n\rangle$ of the form of (10).

By iterating the above procedure, we can generate a set of orthonormal multiphonon states $\{|\alpha_0 = 0\rangle, |\alpha_1\rangle(= |\lambda\rangle), \ldots |\alpha_n\rangle \ldots\}$. This basis is used to diagonalize the residual Hamiltonian, yielding correlated eigenstates

$$|\psi_\nu\rangle = \sum_{\alpha_n} \mathcal{C}^\nu_{\alpha_n} |\alpha_n\rangle \qquad (13)$$

with eigenvalues E_ν.

3. Calculations and results

We applied the EMPM, so reformulated, to ^{16}O. To this purpose, we employed a Hamiltonian composed of an intrinsic kinetic operator and the NN-optimized chiral potential N^{2}LO$_{\rm opt}$ [15].

Figure 1 shows that the convergence properties of the unperturbed ground-state energy do not change when going from the HF to the NAT basis. On the other hand, Fig. 2 shows that, when the NAT basis is used, the point proton radii converge faster with $N_{\max}$ and are insensitive to the HO frequency over a large interval, consistently with Ref. [2]. Large differences emerge once the EMPM is adopted. In fact, we have computed the ground-state energy of ^{16}O in a configuration space including up to 2 phonons. Both HF and NAT bases were generated in the HO space with the maximum oscillator shell $N_{\max} = 12$. The TDA phonons (9) were generated in spaces with $N_{\max}^{\mathrm{TDA}} \leq N_{\max}$.

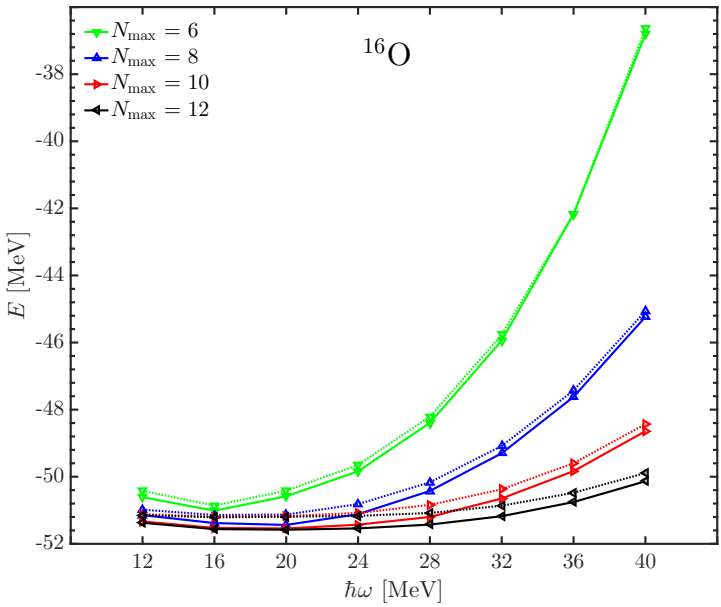

Fig. 1. The HF (full lines) and the NAT (dotted lines) ground-state energies, $E = \langle \mathrm{HF}|\widehat{H}|\mathrm{HF}\rangle$ and $E = \langle \mathrm{NAT}|\widehat{H}|\mathrm{NAT}\rangle$, calculated for various $N_{\max}$, plotted *versus* the HO frequency.

Figure 3 shows that, when natural orbits are used, the total energy E_{tot} converges much faster with $N_{\max}^{\mathrm{TDA}}$ and is practically independent of the HO frequency.

It is also worth noticing that the unperturbed HF ground-state energy is ~ 100 keV lower than the corresponding NAT energy. We have, in fact, for $\hbar\omega = 16.3$ MeV, $E_{\mathrm{HF}} = -51.59$ MeV and $E_{\mathrm{NAT}} = -50.97$ MeV, and for $\hbar\omega = 26$ MeV, $E_{\mathrm{HF}} = -51.48$ MeV and $E_{\mathrm{NAT}} = -50.91$ MeV. This is not surprising, since HF has a variational character and provides the extremal value in the subspace of Slater determinants.

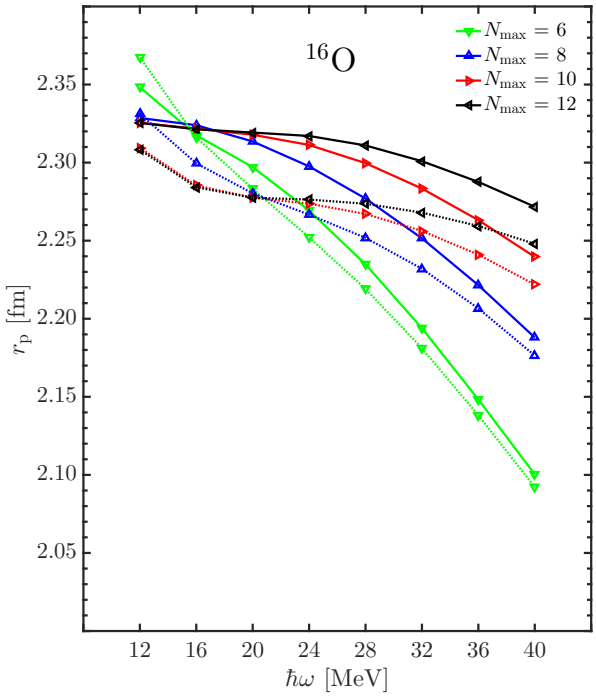

Fig. 2. The proton point-like radii $r_p = \sqrt{\langle \mathrm{HF}|r^2|\mathrm{HF}\rangle}$ (full lines) and $r_p = \sqrt{\langle \mathrm{NAT}|r^2|\mathrm{NAT}\rangle}$ (dotted lines), calculated for different $N_{\max}$, plotted *versus* the HO frequency.

The opposite holds for the correlation energies, as shown in Table I. Thus, the resulting total energies are very close in both bases, with a slight energy gain in favor of the NAT basis. We have, in fact, $E_{\rm tot}^{(\rm HF)} = -108.01$ MeV and $E_{\rm tot}^{(\rm NAT)} = -108.30$ MeV for HO frequency $\hbar\omega = 16.3$ MeV, and $E_{\rm tot}^{(\rm HF)} = -108.32$ MeV and $E_{\rm tot}^{(\rm NAT)} = -108.62$ MeV for HO frequency $\hbar\omega = 26$ MeV.

In both cases, the calculation underestimates the binding energy per nucleon by ~ 1 MeV. This large deviation should be ascribed to the truncation of the space. We expect, in fact, that the inclusion of three and, especially, four phonons will push through their coupling the two-phonon states downward in energy, thereby strengthening the two-phonon coupling to the unperturbed ground state.

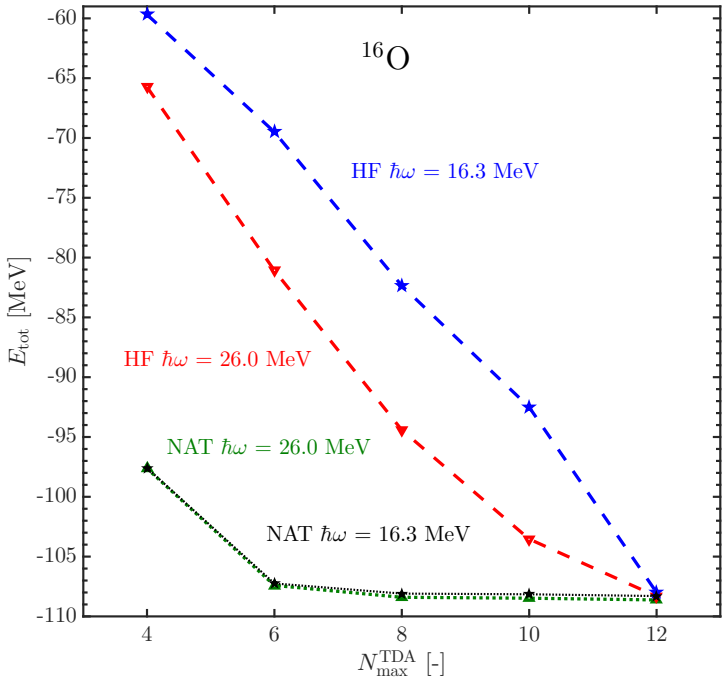

Fig. 3. The total energy $E_{\rm tot}$ of ^{16}O for different values of $N_{\rm max}^{\rm TDA}$. $E_{\rm tot} = E_{\rm HF} + E_{\rm corr}$ for the HF basis (HF) and $E_{\rm tot} = E_{\rm NAT} + E_{\rm corr}$ for the natural orbital basis (NAT). The energies are shown for two values of the HO frequency, $\hbar\omega = 16.3$ MeV and $\hbar\omega = 26$ MeV.

TABLE I

The correlation energies, $E_{\rm corr}$ [MeV], calculated in the HF basis (HF) and the basis of natural orbitals (NAT) for two values of the HO frequency, $\hbar\omega = 16.3$ MeV and $\hbar\omega = 26$ MeV.

$N_{\rm max}^{\rm TDA}$	4	6	8	10	12
(HF) $\hbar\omega = 16.3$ MeV	−8.09	−17.91	−30.76	−40.91	−56.41
(NAT) $\hbar\omega = 16.3$ MeV	−46.62	−56.27	−57.12	−57.18	−57.33
(HF) $\hbar\omega = 26$ MeV	−14.25	−29.60	−42.93	−52.05	−56.84
(NAT) $\hbar\omega = 26$ MeV	−46.72	−56.50	−57.49	−57.57	−57.71

4. Conclusions and outlook

Our analysis has shown that the convergence of the ground-state correlation energy with the space dimensions is much faster if the basis of natural orbitals replaces the HF basis. We will explore in the future if an analogous improvement is achieved for other observables such as the radii and, more general, for all spectroscopic properties like excitation energies and transition strengths.

P.V. and F.K. acknowledge the Czech Science Foundation (grant No. P203-19-14048S). F.K. acknowledges partial support from project UNCE/SCI/013. This work was supported by the Grant Agency of the Czech Technical University in Prague, grant No. SGS19/187/OHK4/3T/14. Computational resources were provided by the CESNET LM2015042 and the CERIT Scientific Cloud LM2015085, under the programme "Projects of Large Research, Development, and Innovations Infrastructures".

REFERENCES

[1] C. Constantinou, "Natural Orbitals for the No-Core Configuration Interction Approach", Ph.D. Thesis, University of Notre Dame, 2017.

[2] A. Tichai, J. Müller, K. Vobig, R. Roth, *Phys. Rev. C* **99**, 034321 (2019).

[3] E.R. Davidson, *Rev. Mod. Phys.* **44**, 451 (1972).

[4] M.R. Strayer, W.H. Bassichis, A.K. Kerman, *Phys. Rev. C* **8**, 1269 (1973).

[5] A. Szabo, N.S. Ostlund, *Modern Quantum Chemistry: Introduction to Advanced Electronic Structure Theory*, Dover Publications Inc., 1996.

[6] F. Andreozzi *et al.*, *Phys. Rev. C* **75**, 044312 (2007).

[7] F. Andreozzi *et al.*, *Phys. Rev. C* **78**, 054308 (2008).

[8] D. Bianco *et al.*, *Phys. Rev. C* **85**, 014313 (2012).

[9] G. De Gregorio *et al.*, *Phys. Rev. C* **95**, 024306 (2017).

[10] F. Knapp *et al.*, *Phys. Rev. C* **92**, 054315 (2015).

[11] G. De Gregorio, F. Knapp, N. Lo Iudice, P. Veselý, *Phys. Rev. C* **93**, 044314 (2016).

[12] G. De Gregorio, F. Knapp, N. Lo Iudice, P. Veselý, *Phys. Rev. C* **94**, 061301(R) (2016).

[13] G. De Gregorio, F. Knapp, N. Lo Iudice, P. Veselý, *Phys. Rev. C* **97**, 034311 (2018).

[14] G. De Gregorio, F. Knapp, N. Lo Iudice, P. Veselý, *Phys. Rev. C* **99**, 014316 (2019).

[15] A. Ekström *et al.*, *Phys. Rev. Lett.* **110**, 192502 (2013).