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A parallel version of ARGOS: A distributed memory model for shared memory UNIX computers*

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Summary. A distributed memory programming model was used in a fully parallel implementation of the *ab initio* integral evaluation program ARGOS (R. Pitzer (1973) *J. Chem. Phys.* 58:3111), on shared memory UNIX computers. The method used is applicable to many similar problems, including derivative integral evaluation. Only a few lines of the existing sequential FORTRAN source required modification. Initial timings on several multi-processor computers are presented. A simplified version of the programming tool used is also presented, and general consideration is given to the parallel implementation of quantum chemistry algorithms.

Key words: Parallel algorithms – Shared memory computers – *Ab initio* algorithms

Introduction

The general application of *ab initio* chemistry methods is still limited by the vast amount of computer resources required to perform electronic structure calculations of any quality on even small molecular systems. For this reason computational chemists have been tracking developments in computer technology, and developing algorithms and programming models appropriately. Notable examples would be the development of matrix based algorithms for vector computers (e.g. the CRAY-1 [1]), the use of local attached array processors (e.g. the FPS-164 [2]), the the use of large memory algorithms [3]. More recently there has been active interest in the exploitation of parallel computers [4–7]. It is noted that nearly all current super- and mini-super-computers use multiple processors

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I have thought it of interest to check the result described in [1] by finding an independent proof based on the "singular value decomposition method" [4] or "pairing theorem" [5, 6]. The alternative proof can be outlined as follows.

The unitary transformations $\boldsymbol{T}$ and $\boldsymbol{U}$ of the basis orbitals induce a similarity transformation $\mathscr{P} \to \mathscr{P}'$ where

$$\mathscr{P}' = \begin{pmatrix} 0 & \boldsymbol{T}^\dagger \boldsymbol{P} \boldsymbol{U} \\ \boldsymbol{U}^\dagger \boldsymbol{P}^\dagger \boldsymbol{T} & 0 \end{pmatrix} = \begin{pmatrix} \boldsymbol{T}^\dagger & 0 \\ 0 & \boldsymbol{U}^\dagger \end{pmatrix} \begin{pmatrix} 0 & \boldsymbol{P} \\ \boldsymbol{P}^\dagger & 0 \end{pmatrix} \begin{pmatrix} \boldsymbol{T} & 0 \\ 0 & \boldsymbol{U} \end{pmatrix}.$$

This similarity transformation does not change the eigenvalues. According to the pairing theorem, we can choose the transformations $\boldsymbol{T}$ and $\boldsymbol{U}$ which enable $\boldsymbol{P}' = \boldsymbol{T}^\dagger \boldsymbol{P} \boldsymbol{U}$ to be diagonal: $\boldsymbol{P}' = (\mathrm{diag}(q_i)\,|\,\boldsymbol{0})$. Then $\mathscr{P}'$ becomes a direct product of 2 by 2 matrices

$$\begin{pmatrix} 0 & q_i \\ q_i^* & 0 \end{pmatrix},$$

the eigenvalues of which are $\varepsilon_{1,2} = \pm\sqrt{|q_i|^2}$. At the same time matrix $\boldsymbol{P}'\boldsymbol{P}'^\dagger = \mathrm{diag}(|q_i|^2)$, so the eigenvalues of $\mathscr{P}'$ and thus of $\mathscr{P}$ are the square roots of those for $\boldsymbol{P}\boldsymbol{P}^\dagger$. This proves the result of [1].

Finally, let me mention the problem that Jug's bond index is of limited interest and applicability, as it cannot be applied at the *ab initio* level of theory (overlapping basis orbitals) since it is not invariant under the non-unitary hybridization transformations that become admitted in that case. One may observe that the sum of the eigenvalues $\lambda_i = |q_i|^2$ (instead of their square roots), gives Wiberg's bond index [7]

$$W_{AB} = \sum_{\mu \in A} \sum_{\nu \in A} |P_{\mu\nu}|^2 = Tr(\boldsymbol{P}\boldsymbol{P}^\dagger) = \mathrm{Tr}(\boldsymbol{P}'\boldsymbol{P}'^\dagger) = \sum_i |q_i|^2.$$

This is a much more useful quantity than Jug's index and also has a proper generalization to the *ab initio* theory [8]. For molecules with no delocalized bonds, the eigenvalues λ_i are close to either 1 or 0, so the numerical values of Wilberg's and Jug's indices should not differ very much in practice.

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