# A Master-worker Type Eigensolver for Large-scale Molecular Orbital Computations

Tetsuya Sakurai[1,3], Yoshihisa Kodaki[1], Hiroto Tadano[1,3], Yuichi Inadomi[2,3], Hiroaki Umeda[2,3], Toshio Watanabe[2,3], and Umpei Nagashima[2,3]

[1] Graduate School of Systems and Information Engineering, University of Tsukuba, Tsukuba 305-8573, Japan
[2] Computational Science Research Division, National Institute of Advanced Industrial Science and Technology
[3] Core Research for Evolutional Science and Technology, Japan Science and Technology Agency

**Abstract.** We consider a parallel method for solving generalized eigenvalue problems that arise from large-scale molecular orbital computations. We use a moment-based method that finds several eigenvalues and their corresponding eigenvectors in a given domain, which is suitable for master-worker type parallel programming models. We also show the efficiency of the Rayleigh-Ritz procedure for improving the accuracy of the results. In addition, we report the performance of the application of the proposed method to several PC clusters connected through a hybrid MPI and GridRPC system.

## 1 Introduction

In the present paper, we consider a parallel method for finding several eigenvalues and eigenvectors of generalized eigenvalue problems in a grid computing environment. A moment-based method [7] to find eigenvalues that lie inside a given domain is used. In this method, a small matrix pencil that has only the desired eigenvalues is derived by solving systems of linear equations. Since these linear systems can be solved independently, we can obtain a master-worker type parallel algorithm. In [8], a parallel implementation of the method using a GridRPC system is presented. We also show the efficiency of the Rayleigh-Ritz procedure for improving the accuracy of the results.

As a test problem, we used the matrices that arise in the calculation of molecular orbitals obtained from the fragment molecular orbital (FMO) method [1]. The FMO method has been proposed as a method for calculating the electronic structure of large molecules such as proteins. The target molecule is divided into small fragments and is suitable for parallel processing. The molecular orbitals of the FMO method are obtained by solving the generalized eigenvalue problem for the Fock matrix calculated with the total density matrix of the FMO method. In this eigenvalue problem, we need several number of eigenvectors related to chemical activities. We report the performance of the application of the proposed method to several PC clusters connected through a hybrid MPI and GridRPC system.

## 2 A master-worker type parallel method

In this section, we briefly describe a master-worker type method for generalized eigenvalue problems presented in [7, 8]. This method, which is based on a moment-based root finding method [2], finds several eigenvalues that are located inside given circles. The error analysis of the method is considered in [3, 6].

Let $A, B \in \mathbb{R}^{n\times n}$, and let $(\lambda_j, \boldsymbol{q}_j)$ $(1 \le j \le n)$ be eigenpairs of the matrix pencil $(A, B)$. For nonzero vectors $\boldsymbol{u}, \boldsymbol{v} \in \mathbb{R}^n$, we define

$$f(z) = \boldsymbol{u}^T(zB - A)^{-1}\boldsymbol{v}$$

with a complex parameter $z$. Then $f(z)$ can be expressed as $f(z) = \sum_{j=1}^{n} \nu_j/(z-\lambda_j)+g(z)$, where $g(z)$ is a polynomial [7]. Let $\Gamma$ be a circle with radius $\rho$ centered at $\gamma$. Suppose that $m$ distinct eigenvalues $\lambda_1, \ldots, \lambda_m$ are located inside $\Gamma$, and let $\omega_j = \gamma + \rho\exp(2\pi\mathrm{i}(j + 1/2)/N)\,(0 \le j \le N - 1)$ with an integer $N$. Let

$$\boldsymbol{y}_j = (\omega_j B - A)^{-1}\boldsymbol{v}, \quad j = 0, 1, \ldots, N - 1, \tag{1}$$

and

$$\boldsymbol{s}_k = \frac{1}{N}\sum_{j=0}^{N-1}(\omega_j - \gamma)^{k+1}\boldsymbol{y}_j, \quad k = 0, 1, \ldots, m - 1. \tag{2}$$

The approximate eigenvalues $\hat{\lambda}_1, \ldots, \hat{\lambda}_m$ and the corresponding approximate eigenvectors $\hat{\boldsymbol{q}}_1, \ldots, \hat{\boldsymbol{q}}_m$ are obtained from $f(\omega_j) = \boldsymbol{u}^T\boldsymbol{y}_j$ $(0 \le j \le N - 1)$ and $\boldsymbol{s}_j$ $(0 \le j \le m-1)$. Approximate residues $\hat{\nu}_1, \ldots, \hat{\nu}_m$ are also obtained. The accuracy of the results is sometimes improved by computing $m'(> m)$ eigenvalues instead of $m$. In this case, we remove $\hat{\lambda}_j$ when $\hat{\lambda}_j$ is located outside of $\Gamma$, or corresponding residue $|\hat{\nu}_j|$ is very small.

In order to evaluate the value of $f(z)$ at $z = \omega_j$, $j = 0, \ldots, N-1$, we solve the Eq. (1). When matrices $A$ and $B$ are large, the computational costs for solving the linear systems are dominant in the method. Since these linear systems are independent, they are solved on remote servers in parallel.

The procedures for solving linear systems are performed on remote servers. Since $A$ and $B$ are common in each system of linear equations, we initially send these data to each server. In order to solve another equation on the same server, a scalar parameter $\omega_j$ is sent. In this approach, we do not need to exchange data between remote servers. Therefore, the present method is suitable for master-worker programming models.

## 3 Modification by the Rayleigh-Ritz procedure

In this section, we show a method for improving the accuracy of the eigenpairs obtained by the method mentioned in the previous section using the Rayleigh-Ritz procedure. Let

$$\hat{Q} = [\hat{\boldsymbol{q}}_1, \hat{\boldsymbol{q}}_2, \ldots, \hat{\boldsymbol{q}}_{m'}].$$

We compute the eigenpairs of $(\theta_j, \boldsymbol{w}_j)$ of the matrix pencil $(\hat{Q}^H A\hat{Q}, \hat{Q}^H B\hat{Q})$. The algorithm is as follows:

**Algorithm**

1. Form $\hat{A} = \hat{Q}^H A \hat{Q}$ and $\hat{B} = \hat{Q}^H B \hat{Q}$.
2. Compute the eigenpairs $(\theta_j, \boldsymbol{w}_j)$ $(1 \leq j \leq m')$ of $(\hat{A}, \hat{B})$.
3. Set $\boldsymbol{x}_j = \hat{Q}\boldsymbol{w}_j$ $(1 \leq j \leq m')$.

The eigenvalue $\theta_j$ of the projected eigenvalue problem $(\hat{A}, \hat{B})$ is taken as approximate eigenvalue of $(A, B)$ with a corresponding approximate eigenvector $\boldsymbol{x}_j = Q\boldsymbol{w}_j$. If the space spanned by the approximate eigenvectors $\hat{\boldsymbol{q}}_1, \ldots, \hat{\boldsymbol{q}}_{m'}$ includes $\boldsymbol{q}_1, \ldots, \boldsymbol{q}_m$ then the Ritz values are good approximations for eigenvalues of $(A, B)$.

## 4 Numerical Examples

We present a numerical example of the proposed method. In order to evaluate the performance of the method for the situation in which several PC clusters are employed via a wide area network, we regarded some parts of a large-size PC cluster system as distributed PC clusters.

Experiments were performed on the AIST F32 Super Cluster of the National Institute of Advanced Industrial Science and Technology. The node of the cluster system was a 3.06-GHz Xeon with 2 GB of RAM. The client machine was a 3.0-GHz Pentium 4 with 2 GB of RAM. The client was connected to servers via Gigabit Ethernet. The program was implemented with a GridRPC system, Ninf-G ver. 2.4.0 [5].

The test matrices were derived from computation of the molecular orbitals of lysozyme (129 amino-acid residues, 1,961 atoms) with 20,758 basis functions. The structure of the lysozyme molecule has been determined experimentally, and we added counter-ions and water molecules around the lysozyme molecule in order to simulate in vivo conditions. The size of both $A$ and $B$ was $n = 20,758$, and the number of nonzero elements was $10,010,416$. $A$ was symmetric, and $B$ was symmetric positive definite.

Since the matrix $\omega_j B - A$ with complex $\omega_j$ is complex symmetric, the COCG method [9] with incomplete Cholesky factorization with a complex shift [4] was used. We used the same precondition matrix in each process of Ninf-G. The stopping criterion for the relative residual was $10^{-12}$. Computation was performed in double-precision arithmetic. The elements of $\boldsymbol{u}$ and $\boldsymbol{v}$ were distributed randomly on the interval $[0, 1]$ by a random number generator.

The intervals $[-0.24, -0.18]$ and $[0.16, 0.22]$ were covered by eight circles. These intervals include the energy levels of the highest occupied molecular orbitals (HOMO) and the lowest unoccupied molecular orbitals (LUMO), which are key factors in the amount of energy needed to add or remove electrons in a molecule. The parameters were chosen as $N = 48$ and $m' = 20$.

We observed the wall-clock time in seconds with various combinations of the numbers of Ninf-G processes and MPI processes. The time required to load the input matrices into the memory of the client and that required to start up the Ninf-G and MPI processes were not included. The results are listed in Table 1.

**Table 1.** Wall-clock times (seconds)

| | | # of Ninf-G processes | | | |
|---|---|---|---|---|---|
| | | 1 | 2 | 4 | 8 |
| # of MPI processes | 1 | 1351 | 670 | 351 | 240 |
| | 2 | 901 | 447 | 238 | 159 |
| | 4 | 529 | 265 | 142 | 82 |
| | 8 | 348 | 173 | 86 | 50 |

Twenty eigenvalues and corresponding eigenvectors were obtained in 50 seconds with the combination of eight Ninf-G processes and eight MPI processes (64 processors). In these computations, the preconditioning matrix was computed in serial code, and was computed once in each Ninf-G process. This causes the load imbalance between MPI processes. The use of a parallel preconditioner will improve the performance.

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