Multiscale Algorithms for Eigenvalue Problems

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Abstract

Iterative multiscale methods for electronic structure calculations offer several advantages for large-scale problems. Here we examine a nonlinear full approximation scheme (FAS) multigrid method for solving fixed potential and self-consistent eigenvalue problems. In principle, the expensive orthogonalization and Ritz projection operations can be moved to coarse levels, thus substantially reducing the overall computational expense. Results of the nonlinear multiscale approach are presented for simple fixed potential problems and for self-consistent pseudopotential calculations on large molecules. It is shown that, while excellent efficiencies can be obtained for problems with small numbers of states or well-defined eigenvalue cluster structure, the algorithm in its original form stalls for large-molecule problems with tens of occupied levels. Work is in progress to attempt to alleviate those difficulties.

Introduction

Electronic structure methods for large-scale problems can be divided into three general categories: plane-wave [1], traditional basis set [2, 3], and real-space methods [4]. Real-space methods result in a banded Hamiltonian, through finite difference [5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17], finite element [18, 19, 20], or wavelet [21] representations of the Laplacian operator. Gaussian basis sets lead to a smaller total matrix size of the Hamiltonian (relative to real-space methods), but the matrix is less banded. The plane-wave basis set, on the other hand, is completely delocalized in real space. The bandedness of the Hamiltonian in real-space methods is advantageous in several respects. First, it leads to ease of developing parallel codes. Second, methods developed recently which scale linearly with the system size generally rely on localization of the orbitals and real-space methods ‘mesh’ well with those algorithms [22]. Third, efficient multiscale methods accelerate convergence by decimating errors over a wide range of length scales. Fourth, finite clusters or periodic systems can be treated with equal effort. Finally, local mesh refinements can be incorporated without
degrading the efficiency of the solver. Several groups have developed real-space solvers for the Kohn-Sham equations of density functional theory (DFT) in the last decade. Multigrid (MG) methods have been employed extensively to accelerate the convergence rate.

A central feature of the Kohn-Sham problem is its nonlinearity. It is nonlinear in two respects. First, the eigenvalue problem itself is nonlinear since one solves for both the eigenvalues and eigenfunctions. Second, the self-consistent potential depends nonlinearly on the charge density obtained from the squares of the eigenstates. Therefore, it can be expected that nonlinear multigrid methods will lead to increased efficiencies. This has indeed been observed previously in studies which compare a full approximation scheme (FAS) nonlinear approach to linearized MG methods.

The most costly operation in the nonlinear eigenvalue approach of Brandt et al. is the Ritz projection (preceded by Gram-Schmidt orthogonalization) on the fine level. If \( q \) orbitals span the whole physical domain, with \( N_h \) grid points on the fine level (labelled by \( h \)), this step of the algorithm scales as \( q^2 N_h^2 \). In the present paper, we carry the nonlinear FAS scheme further by implementing an extension of the Brandt et al. algorithm proposed by Costiner and Ta’asan. In their approach, the orthogonalization and Ritz projection are moved to coarse levels within the FAS strategy. This results in an 8-fold decrease in computational cost for the Gram-Schmidt/Ritz operation for each coarser level in three dimensions. We discuss relevant details of their method, and then present numerical results on fixed potential and self-consistent eigenvalue problems related to atomic and molecular structure.

### Algorithms

Multigrid methods accelerate the convergence rate of iterative relaxation solvers for partial differential equations by decimating errors on a wide range of length scales; it is the long wavelength modes of the error which degrade single-level relaxation efficiency. Nonlinear multigrid methods incorporate a full representation of the problem on coarse levels. Modifications of the fine grid matrix equation are necessary on the coarse grids to obtain zero correction at convergence. These modifications are termed defect corrections. In this paper, we examine the FAS multigrid method of Costiner and Ta’asan. They presented a detailed account of the algorithm in two papers: the first concerns fixed potential problems and the second addresses self-consistency. A brief summary will be given here. We will follow the notation from their work.

**FAS Eigenvalue Method**

Suppose we wish to solve an eigenvalue problem represented in real space with finite differences. This leads to the matrix equation
The matrix $A$ is the $N_h^g \times N_h^g$ (banded) Hamiltonian, $U$ is the $q \times N_h^g$ matrix of the eigenvectors, and $\Lambda$ is the diagonal $q \times q$ matrix of eigenvalues.

In the FAS approach, we express the coarse grid (level $i$) problem as

$$F_i U_i = T_i.$$  \hfill (2)

Here $F_i U_i = A_i U_i - U_i \Lambda_i$ and $T_i$ is the defect correction. On the finest grid $T_i = 0$. On grids $j$ coarser than $i$,

$$T_j = I_j^i (T_i - F_i U_i) + F_j I_j^i U_i,$$  \hfill (3)

where the operator $I_j^i$ is the restriction operator. We use full weighting restriction throughout, which just involves a local trapezoid-rule integration of the function values from the fine grid. In the above equations, if the exact numerical fine grid solution is inserted into the coarse grid equations, identities are obtained. This is equivalent to the zero correction at convergence condition.

An initial approximation is first obtained on the fine grid. We obtain this approximation by implementing a full multigrid (FMG) cycle \[9\], beginning on the coarsest level, interpolating to the next finer grid, performing MG cycles there, and so on until the finest grid is reached. In this way, a good initial approximation is obtained for very little numerical effort. Following relaxation (typically 2-5 Gauss-Seidel or successive over-relaxation/SOR steps), the fine grid approximation is passed to the coarse level by restricting the eigenfunctions, the potential, and the defect correction from the finer grid. Relaxations (and generalized Ritz projections, see below) are performed on the current coarse grid, and the problem is then passed again to the next coarser level. This process is repeated until the coarsest grid is reached. We typically utilize three grid levels when the finest grid is a $65^3$ mesh. Once relaxation is done on the coarsest level, a correction step for the next finer level is performed:

$$U_{i}^{\text{new}} = U_{i}^{\text{old}} + I_j^i (U_j - I_j^i U_{i}^{\text{old}}).$$  \hfill (4)

$I_j^i$ is the interpolation operator. Linear interpolation by lines is used throughout except during the FMG process when passing to the next finer level, where cubic interpolation by lines is used (to obtain a better initial guess on the fine grid). The correction steps are continued until the finest grid functions are corrected followed by relaxation steps there. The full cycle through all the levels is termed a V-cycle. One may then repeat the MG V-cycles until a desired convergence is obtained.

**Generalized Ritz Projection, GRP**

Consider a new eigenvalue relation, in which the matrix $V$ results from a linear combination of the current approximation $U$:
\[ AV = V\Lambda, \]  
(5)

where \( V = UE \) and \( E \) is a \( q \times q \) matrix of normalized vectors, the columns of which determine the coefficients for the linear combination of old vectors from \( U \). Then on the fine grid we have the relation

\[ AUE = UE\Lambda. \]  
(6)

When this problem is passed to the coarser levels, the proper FAS transfer is

\[ AUE = UE\Lambda + TE. \]  
(7)

If we multiply on the left by \( U^T \), we obtain the following generalized (nonsymmetric) eigenvalue problem on the coarse grid:

\[ U^T(AU - T)E = (U^TU)E\Lambda. \]  
(8)

Notice that the eigenfunctions are no longer orthonormal when passed to the coarse grid. Also, it is easy to show that, at convergence, the eigenvalues are the same on all grid levels; in principle, there is no need to compute them on the finest grid. We solve this \( q \times q \) eigenproblem with standard linear algebra packages. Once solved, we obtain new eigenvalues, eigenfunctions (linear combinations of the current approximation), and defect corrections (also linear combinations of the old defect corrections). On the finest grid, where the defect correction \( T \) is zero, the GRP reduces to the usual Ritz projection employed in the Brandt et al. algorithm [24].

**Backrotation, BR**

A subtle aspect of the correction scheme outlined above is that the coarse-grid eigenfunctions must properly match their fine-grid counterparts for the correction step. Therefore, Costiner and Ta’asan [25] introduced a process called backrotation in their solver. This operation is designed to prevent rotations in degenerate or near-degenerate subspaces, and to prevent sign changes, rescalings, and permutations of the eigenvectors. In the backrotation, the \( E \) matrix is modified towards the ends listed above. If this step is not employed in the algorithm of Ref. [25], the solver typically does not fully converge. As a simple example, imagine that the sign of one of the eigenfunctions changes during GRP on a coarse level. If the correction is then interpolated to the fine grid, the corrected function will be severely distorted. At convergence, the \( E \) matrix should approach the unit matrix. During processing (prior to backrotation), it tends to have block diagonal form, where the blocks correspond to degenerate or near-degenerate subspaces. The dimensions of the blocks determine the eigenvalue cluster sizes. An extensive discussion of the backrotation is given in the original paper.

**Relaxation**
A major feature of MG methods is that relatively simple relaxation strategies can be employed so long as they decimate errors with wavelengths comparable to the grid spacing on a given level. Gauss-Seidel is the most common one utilized. We have investigated several relaxation strategies for smoothing on each level. Originally, we used the Gauss-Seidel with shift form given in Ref. [24]. Recently, we have extended this relaxation method to an SOR form, and find improved convergence. On the coarsest level, we have employed Gauss-Seidel directly, Gauss-Seidel with constraints designed to maintain eigenfunction orthonormality on the fine grid [21], and Kaczmarz relaxation. Kaczmarz relaxation is guaranteed to converge, but it exhibits slower convergence relative to Gauss-Seidel or SOR (this is not a significant issue on coarse levels). It will be specified below which method was used for each application. Further details of relaxation methods will be presented in an extensive account of our algorithm [29].

**Self-Consistent Problems**

Some of the applications presented below concern self-consistent solution of the Kohn-Sham equations. In the work presented here, we update the eigenfunctions and self-consistent potential sequentially. That is, given an initial approximation to the effective potential, an MG V-cycle is performed to update the eigenfunctions. From the new eigenfunctions, a new charge density is computed, from which a new effective potential is obtained. The main computational step for updating the effective potential is solution of a Poisson problem. This equation is also solved with MG V-cycles. The total time to solve the Poisson equation is less than that for updating a single eigenfunction, and this operation scales linearly with system size. We are currently exploring approaches to update the effective potential on coarse levels simultaneously with the eigenfunctions [26]. This can be expected to accelerate the self-consistent convergence rate. We note that in our calculations so far, we have found no need for potential or charge density mixing of old and new solutions; we believe this is due to long-wavelength stabilization of the charge density during the FMG preconditioning process.

**Pseudopotentials**

For our calculations on atoms and molecules, we have incorporated the separable dual-space Gaussian pseudopotentials developed by Goedecker *et al.* [30, 31]. These pseudopotentials have analytic forms with only several parameters per atom, and they exhibit optimal decay properties in both real and reciprocal space. We have implemented the real-space relativistic version of these pseudopotentials for the present calculations. For calculations on the coarse grids, we simply compute the function values just as we do on the fine level. Of course fewer grid points are necessary to sample the pseudopotential on coarse grids due to the decay properties of the projectors. It has been shown that application of pseudopotentials in real space is more efficient than in reciprocal space [32].
Numerical Results

Fixed Potential Problems

We first present results of the FAS algorithm on fixed potential problems. As a benchmark, we utilized the original algorithm of Ref. [24] and solved the same two dimensional eigenvalue problem addressed in that paper. Their kinetic energy operator is scaled by a factor of two. A second order finite difference approximation was assumed for the Laplacian. Half of their potential is

\[ v = 5y \sin(3\pi x). \]  

(9)

The total domain size was taken as one, and four grid levels were utilized in the FAS process. Gauss-Seidel relaxation (with a shift parameter of zero) was employed on all four levels. One relaxation step was performed on the coarsest level while enforcing constraints designed to maintain eigenfunction orthonormality on the finest level. On all other levels, two relaxation steps were performed. In total, 7 FAS V-cycles were implemented on the finest level. The computed eigenvalues and residuals are shown in Table 1. The residual for each eigenfunction is defined as

\[ r = \sqrt{\frac{\sum |H\psi - E\psi|^2}{N^h_g}}, \]  

(10)

where the sum is over all the fine grid points and \( N^h_g \) is the total number of grid points. The algorithm with Ritz projection performed on the finest level converges nicely to the numerically exact eigenfunction/eigenvalue pairs.

We next compare the convergence rates of three FAS algorithms: 1) Ritz on fine grid (Ref. [24], 2) generalized Ritz with backrotation (GRBR) on coarse grids with no orthonormalization on the fine level [25], and 3) GRBR on coarse grids with periodic (every 5 V-cycles) Gram-Schmidt orthonormalization on the fine grid. These approaches were tested on simple fixed potential problems, namely the three-dimensional harmonic oscillator and the hydrogen atom. Both of these physical problems have degenerate subspaces leading to eigenvalue clusters which must be handled in the backrotation step. For these problems, Gauss-Seidel relaxation was employed on all levels, except Kaczmarz relaxation was utilized for the coarsest grid relaxation for the hydrogen atom problem (this led to increased efficiency).

The convergence results are presented in Figs. 1 and 2. For the harmonic oscillator problem, we solved for ten states and used a 12th-order finite difference representation for the Laplacian. The total domain size is ten, and the fine grid is a \( 65^3 \) mesh; three grid levels were utilized. Atomic units are used throughout. A shift parameter equal to the current eigenvalue was employed in the Gauss-Seidel relaxations (5 steps per level). The
GRBR step was performed on the coarsest level. A total of 15 V-cycles were conducted. The compute times per V-cycle for the three algorithms listed above were 50.9, 34.6, and 36.6 sec., respectively, on an 800 MHz machine. The $q$ values are small enough that the orthogonalization and Ritz projections do not yet dominate the compute time. For the hydrogen atom case, we generated the fixed potential by numerical solution of the Poisson equation for a fixed central unit charge. Again, 12th-order finite differences were employed, and we solved for 14 states on a domain with a total side length of 28. The fine grid is a $65^3$ mesh. For this case, it was found that a shift parameter equal to the potential was more efficient. A total of 20 V-cycles were conducted. For both cases, the fastest (lowest eigenvalue) and slowest (highest eigenvalue) converging cases are shown. The GRBR (without orthonormalization) operation was conducted on the middle grid level, while for the third algorithm with periodic Gram-Schmidt operations on the fine grid, the GRBR step was implemented on the coarsest level. The compute times per V-cycle were 81.5, 59.7, and 52.8 sec. for the three algorithms.

For both physical problems, performing the orthonormalization and Ritz projection on the fine level leads to the most efficient convergence. The harmonic oscillator potential is smooth, and the GRBR algorithm exhibits good convergence behavior. However, the convergence rate is slightly slower than when orthonormalization and Ritz projection are performed on the finest level. The GRBR convergence rate is the same whether or not orthonormalization is periodically done on the finest level, indicating good separation can be obtained without processes on the fine level. For the hydrogen atom case, the overall convergence rate is reduced relative to the harmonic oscillator, presumably due to the singular nature of the potential. Also, periodic Gram-Schmidt operations on the finest level increase the convergence rate slightly.

As part of the backrotation step, the degenerate subspaces (eigenvalue clusters) must be identified so as to prevent rotations within those clusters. Both the harmonic oscillator and the hydrogen atom problems possess clear eigenvalue structure which can be directly incorporated or determined during the solution process. This ensures healthy convergence of the GRBR algorithm, behavior which was also observed in the original work of Costiner and Ta’asan [25]. We will see below that, for large self-consistent molecular cases without such well-defined eigenvalue cluster structure, the algorithm may stall due to mixing during the backrotation step.

Self-Consistent Pseudopotential Calculations

Computational results for self-consistent pseudopotential Schrödinger-Poisson eigenvalue problems are presented. Just as for the fixed potential problems presented above, the algorithms used were Ritz projection on the fine level coupled with Gram-Schmidt orthogonalisation and the GRBR algorithm without and with periodic fine grid Gram-Schmidt operations. In both GRBR algorithms, the eigenfunctions were normalized on the fine level to ensure charge conservation. Three examples, Ne, CO, and the benzene dithiol molecule were used in the study. All three cases are three dimensional and were treated with 12th
order finite difference representations. Three grid levels were utilized comprising $17^3$, $33^3$ and $65^3$ total points. The first example, the Ne atom, possesses 4 occupied states. Similarly 5 and 21 eigenvectors were required for the CO and benzene dithiol molecules, respectively. The Ne and CO examples possess well-defined eigenvalue cluster structure (triply degenerate p states for Ne and a doubly degenerate $\pi$ bonding orbital for CO). Convergence results are presented in Figs. 3-5.

Choosing the optimal parameters for the relaxation scheme was important. The shift parameter for Gauss-Seidel relaxation was taken as $\lambda + v$, where $\lambda$ is the eigenvalue and $v$ is symbolically the effective potential (nonlocal in the case of the pseudopotential). Gauss-Seidel relaxation ($\omega = 1$, where $\omega$ is the overrelaxation parameter) was used on all levels except on the finest grid where SOR relaxation was employed ($\omega = 1.7$). These near-optimal relaxation parameters were determined by numerical experiments. In the final V-cycle involving the three grid levels, three relaxation steps were done on each level.

All three algorithms converged for the Ne and CO cases. The fine grid Ritz plus Gram-Schmidt and coarse-grid GRBR with periodic fine level Gram-Schmidt algorithms both exhibited excellent convergence rates. Implementing GRBR with no fine grid separation (orthogonalization) led to slower convergence; the total energy convergence slows after a few self-consistency iterations. In the GRP, the fine level separation of wavefunctions comes as a result of GRBR done on the middle level. But complete separation may not always be achieved on the fine level (Table 2). Performing only a few fine grid Gram-Schmidt operations during the entire solution process restores the convergence rate and leads to acceptable fine grid eigenfunction orthogonality at the end. Clearly the form of the potential (self-consistent pseudopotential in this case) and resulting eigenvalue structure have impacts on the convergence behavior of the GRBR algorithm.

In the case of benzene dithiol, the fine grid separation algorithm converged nicely as for the smaller molecules. However, neither variant of the GRBR algorithm converged properly; the solver stalled with only modest energy convergence. To probe for the reason for this lack of convergence, we first converged the system using fine-grid separation for 15 V-cycles, and then used the resulting potential in a fixed potential calculation. We observed that the $E$ matrix determined from GRBR gradually begins to destabilize rather than to converge to the unit matrix as expected. This suggests again that the form of the potential and the resulting eigenvalue cluster structure affect the convergence of the GRBR algorithms. Benzene dithiol is a relatively large molecule with 14 atoms (C,O,S and H) and 21 valence eigenstates. We believe that the difficulties arise from the ambiguous cluster structure of the eigenvalues which leads to mixing during the backrotation operation. Similar calculations were performed on the benzene molecule, which possesses clear symmetry and cluster structure, and the algorithm converged. Since determination of the cluster structure is crucial for convergence, and this must be performed automatically in the algorithm in order to treat general systems, this issue must be addressed for the GRBR approach to provide a generally convergent scheme. Work is in progress investigating these difficulties. One possible solution is suggested below.
Discussion

The objective of this paper has been to provide a test of the nonlinear FAS multigrid eigenvalue method of Refs. \[25, 26\] for solving the Kohn-Sham equations. This method is promising because it removes the expensive orthogonalization and Ritz projection operations to coarse levels (with a corresponding 8-fold reduction in cost per level in three dimensions). The necessary fine grid work only involves relaxation, normalization of the eigenfunctions, and perhaps orthogonalization within degenerate clusters. The model problems treated in Refs. \[25, 26\] possess relatively smooth potentials and well-defined eigenvalue cluster structure. Similar to the results of Costiner and Ta’asan, we find good convergence of the GRBR approach for fixed potential and self-consistent eigenvalue problems with well-defined eigenvalue clusters. However, we found that for a larger molecular case with complicated eigenvalue structure, the GRBR approach did not converge properly. We linked these difficulties to the backrotation step which appears to be highly sensitive to the determination of the clusters. Since determination of the clusters must be done numerically during the course of the solution process, this issue must be addressed to develop a generally convergent solver for large systems.

We have recently begun investigating one possible approach to deal with these difficulties. Notice that the $E$ matrix of Eqs. 6 and 7 is formally the same on all levels at convergence (just as are the eigenvalues). In the algorithm of Ref. \[25\], the backrotation involves a modification of the $E$ matrix designed to prevent rotations in degenerate or near-degenerate subspaces, sign changes, rescalings, and permutations of eigenvectors. Utilizing the fact that the $E$ matrix is the same on the coarse and fine levels (at convergence), we can circumvent the backrotation by first applying the $E$ matrix to the current fine-grid approximation to the eigenfunctions prior to the correction step. This approach is in a sense a hybrid of the fine and coarse grid Ritz projections; we use the coarse grid to generate the new eigenvalues and the $E$ matrix, but we use that $E$ matrix to alter the fine-grid occupied subspace. Thus the expensive step of constructing the Ritz matrix has been moved to the coarser level. The use of the $E$ matrix to update the fine-grid function is a relatively cheap operation. Formally, it does scale as $q^2 N_q^h$, but the $E$ matrix is of block diagonal form, with the blocks of dimension of the corresponding degenerate cluster. These clusters are generally very small. Therefore realistically the update operation scales as $q N_q^h$ if the eigenfunctions span the whole domain. Some discussion along these lines was already given in Ref. \[24\]. We are currently exploring the use of this idea in our nonlinear FAS multigrid solver. In preliminary results, we have found it to converge properly for all of the physical problems examined in this paper.

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| No | $2\lambda$     | $r$   | $\lambda$     |
|----|----------------|-------|----------------|
| 1  | 18.71847149    | 3.3E−11 | 18.71847149   |
| 2  | 48.18927363    | 4.1E−09 | 48.18927363   |
| 3  | 51.56004355    | 5.0E−12 | 51.56004355   |
| 4  | 81.07201016    | 2.8E−11 | 81.07201016   |
| 5  | 97.00117915    | 9.6E−09 | 97.00117915   |
| 6  | 99.57484220    | 1.2E−08 | 99.57484220   |
| 7  | 129.1084354    | 1.7E−06 | 129.1084354   |
| 8  | 129.8996943    | 2.5E−07 | 129.8996943   |

Table 1: Comparison of eigenvalues to results of Ref. [24].
Table 2: A sample of dot products of wavefunctions of CO and Ne are shown. Column GS includes products when the Gram-Schmidt orthogonalization was performed and it serves only as a reference. The column GRBR includes the products as a result of GRBR only and the wavefunctions are not fully separated. The column GRBR+GS includes the products when Gram-Schmidt orthogonalization was performed at a regular interval (5,10,15 V-cycles only). The separation of wavefunctions is improved. The total number of V-cycles performed was 20.
Figure 1: Convergence rates for different methods in a fixed harmonic oscillator potential. Long dashed lines are the results of the Ritz method on the fine grid, the solid lines are results of the GRBR method on the coarsest level, and the dotted lines with diamonds are results of the GRBR method on the coarsest level with additional periodic Gram-Schmidt orthogonalization on the fine grid.
Figure 2: Convergence rates for different methods in a fixed hydrogen atom potential. Long dashed lines are the results of the Ritz method on the fine grid, the solid lines are the results of the GRBR method on the $33^3$ grid, and the dotted lines with diamonds are results of the GRBR method on the $17^3$ grid with additional periodic Gram-Schmidt orthogonalization on the fine grid.
Figure 3: Convergence rate for Ne. The logarithm (base 10) of the difference between the current and fully converged total energies is plotted against the number of V-cycles (self-consistency steps). Ritz and GRBR stand for fine-grid Ritz projection and coarse-grid generalised Ritz projection, respectively. GRBR-GS is for GRBR with fine-grid Gram-Schmidt orthogonalization performed at 3 V-cycles (5,10,15). The fine grid spacing used was $h=0.178437$. 
Figure 4: Convergence rate for CO. The logarithm (base 10) of the difference between the current and fully converged total energies is plotted against the number of V-cycles (self-consistency iterations). Ritz and GRBR stand for fine grid Ritz projection and coarse-grid generalized Ritz projection with backrotation, respectively. GRBR-GS is for coarse-grid GRBR with Gram-Schmidt orthogonalization performed on the fine grid at 3 V-cycles (5,10,15). The fine grid spacing used was h=0.178437.
Figure 5: Convergence rate for benzene dithiol. The logarithm (base 10) of the difference between the current total energy and the fully converged value is plotted against the number of V-cycles (self-consistency iterations). Only the fine grid Ritz projection case is shown. The fine grid spacing used was $h=0.3$. 

![Graph showing convergence rate](image)