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A multilevel correction adaptive finite element method for Kohn–Sham equation ☆

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ABSTRACT

In this paper, an adaptive finite element method is proposed for solving Kohn–Sham equation with the multilevel correction technique. In the method, the Kohn–Sham equation is solved on a fixed and appropriately coarse mesh with the finite element method in which the finite element space is kept improving by solving the derived boundary value problems on a series of adaptively and successively refined meshes. A main feature of the method is that solving large scale Kohn–Sham system is avoided effectively, and solving the derived boundary value problems can be handled efficiently by classical methods such as the multigrid method. Hence, the significant acceleration can be obtained on solving Kohn–Sham equation with the proposed multilevel correction technique. The performance of the method is examined by a variety of numerical experiments.

1. Introduction

Density functional theory (DFT) is one of the most successful models in the quantum mechanics. It breaks the efficiency bottleneck in the study of manybody system caused by the high dimensionality, and has been playing a crucial role in the study of the Schrödinger equation, theoretically and numerically. To date, DFT has been widely used in a variety of areas such as computational quantum chemistry, nano material, and quantum optics. Lots of innovative techniques have been developing with DFT.

Among those models in DFT, Kohn–Sham model [15] attracts more and more attention due to its good ability on balancing the accuracy and the efficiency, compared with other models such as orbital-free DFT [6,9]. There have been lots of numerical methods available in the market. For instance, plane-wave method [31] is the most popular method in the computational quantum chemistry community. Owing to the independence of the basis function to the ionic position, plane-wave method has advantage on calculating intermolecular force. Combined with the pseudopotential method, plane-wave method plays an important role in the study of the ground and excited states calculations, and geometry optimization of the elec-

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tronic structures. Although plane-wave method is popular, there are several drawbacks since their natural requirements on the regular shape of the domain, and periodic boundary conditions. To make the numerical methods more suitable for the practical calculations, the real space methods have been developing to solve the Kohn–Sham equation. For instance, finite difference methods [2], finite element methods [23], discontinuous Galerkin methods [16], wavelet methods [13], etc. For these methods, fast Fourier transform can be avoided in the calculation, and then the unstructured meshes can be employed and the domain with complicated geometry can be handled in a nature way. Since the local basis functions are used to represent the wavefunction, these methods are quite flexible to handle different boundary conditions. Furthermore, the locality of the basis function results in the sparsity of the system. Hence, a number of mature fast algorithms such as the multigrid method can be employed to accelerate the algorithm. Recently, the adaptive methods are introduced in solving Kohn–Sham equation to further improve the algorithm efficiency.

In a typical adaptive method for solving Kohn–Sham equation, the algorithm consists of the following process, i.e.,

$$\ldots \textbf{Solve} \rightarrow \textbf{Estimate} \rightarrow \textbf{Mark} \rightarrow \textbf{Refine and/or Coarsen} \ldots .$$

In **Solve** step, the Kohn–Sham equation is solved on the current mesh. Then an error indicator is assigned for each element in the mesh with certain error estimation technique in **Estimate** step. In **Mark** step, every element in the current mesh is marked for refining or coarsening with certain marking strategy. In the last step, the local refinement and/or coarsening is implemented on the current mesh to generate a new nonuniform mesh, and then a new round adaptive process is launched. The above adaptive process is employed in several works related to the adaptive numerical methods for Kohn–Sham equation [3,8,21], and improvement on the simulation efficiency can be observed successfully. To further improve the implementation efficiency, there are two traditional ways to follow. The first one is to develop a more accurate error estimation method. With more accurate error indicator, the mesh can be locally refined and/or coarsened more precisely, which means that total number of the mesh elements can be controlled effectively to reach almost the same accuracy of the numerical solution. In the second way, people may resort to the parallel computing. Since its nonlinearity, certain iteration needs to be introduced to linearize the Kohn–Sham equation, which means that a series of linear and generalized eigenvalue problems needs to be solved for a given mesh. Hence, it is important to control the size of the discretized system for the acceleration of the simulations. More importantly, to preserve the orthogonality among those wavefunctions, the computational complexity for the orthogonalization is $\mathcal{O}(N_g N_w^2)$, where N_g and N_w are the numbers of the mesh elements and the wavefunctions, respectively. It is clear that when the size of the electronic structure system, i.e., N_w is small enough, the computational complexity roughly depends on the size of the discretized system, i.e., N_g, linearly. However, when N_w becomes nonignorably large, N_w^2 would dominate the computational complexity. In this case, more efficient algorithm on preserving the orthogonality of the wavefunctions is needed, or the issue can be partially resolved by effectively control N_g since when N_w is large, a sufficiently large N_g is needed for the quality spatial discretization. Consequently, it is crucial to control the size of the discretized Kohn–Sham equation for a given electronic structure system, from the efficiency point of view.

In [11,14,17,27–30], Xie and his co-workers proposed and developed a multilevel correction technique towards controlling the size of the discretized nonlinear eigenvalue problems. With this technique, the nonlinear eigenvalue problem is always discretized and solved on a fixed and moderately coarse mesh with the finite element method whose basis functions are improved by a series of boundary value problems defined on a successively and adaptively refined meshes. The advantage of the proposed multilevel correction technique is twofold. First, the mesh and the number of the degree of freedoms for discretizing the nonlinear eigenvalue problem are fixed during the whole simulation. In other words, the discretized size of the nonlinear system is fixed during the whole simulation. The efficiency and required memory of the simulation will benefit from this property a lot if a quality eigensolver is available. Second, although a series of boundary value problems with increasing sizes need to be solved, there are lots of mature methods such as the multigrid method to make the solving efficient. With the above two advantages, the size issue for solving the nonlinear eigenvalue problems is partially resolved, and excellent results can be observed from [11,14,17,28–30].

Following the idea of the multilevel correction, an adaptive finite element method is developed in this paper for solving Kohn–Sham equation. The method can be described as follows. Initially, a moderately coarse mesh is prepared for the Kohn–Sham system. The size of this coarse mesh depends on the given electronic structure system. After solving the Kohn–Sham equation with the self-consistent field iteration (SCF) method [24,25], the mesh will be adaptively refined and/or coarsened with the error indicator generated by some error estimation method. By solving a linear boundary value problem derived from plugging the approximate solutions into the Hamiltonian and right hand side of the Kohn–Sham equation, the new approximation for the wavefunctions are obtained. It is analyzed in this paper that this procedure can improve the accuracy of approximate wavefunctions effectively. Hence, it is reasonable to use these more accurate wavefunctions to enrich the previous coarse finite element space for the Kohn–Sham equation. Then a new round iteration is launched till some stop criterion is reached. It is noted that the mesh as well as the number of the degree of freedoms for the discretization of the Kohn–Sham system are not change during the whole simulation. In the Hamiltonian of the Kohn–Sham equation, the full potential expression is employed for the external potential term, and local density approximation is used. In the implementation, the generalized eigenvalue problem is solve by the implicitly restarted Lanczos method in the package ARPACK, while the geometric multigrid (GMG) method is used to solve the boundary value problem. The effectiveness on accelerating the adaptive finite element method for solving Kohn–Sham equation is successfully shown by a variety of numerical experiments.