East Asian Journal on Applied Mathematics
doi: 10.4208/eajam.2022-134.081022

Vol. **13**, No. 2, pp. 299-319
May 2023

A Linearized Structure-Preserving Numerical Scheme for a Gradient Flow Model of the Kohn-Sham Density Functional Theory

Guanghui Hu[1,2,3], Ting Wang[1] and Jie Zhou [4,*]

[1]*Department of Mathematics, Faculty of Science and Technology, University of Macau, Macao SAR, China.*
[2]*Zhuhai UM Science & Technology Research Institute, Zhuhai, Guangdong Province, China.*
[3]*Guangdong-Hong Kong-Macao Joint Laboratory for Data-Driven Fluid Mechanics and Engineering Applications, University of Macau, Macao SAR, China.*
[4]*School of Mathematical and Computational Sciences, Xiangtan University, Xiangtan, Hunan Province, China.*

Received 22 April 2022; Accepted (in revised version) 8 October 2022.

Abstract. Dai *et al.* [Multiscale Model. Simul. **18** (2020)] proposed a gradient flow model and a numerical scheme for ground state calculations in Kohn-Sham density functional theory. It is a feature that orthonormality of all wave functions can be preserved automatically during the simulation which makes such a method attractive towards simulations for large scale systems. In this paper, two extensions are proposed for further improving the efficiency of the method. The first one is a linearization of the original nonlinear scheme. It is shown analytically that both the orthonormality of wave functions and the decay of the total energy can be preserved well by this linear scheme, while a significant acceleration can be observed from the numerical experiments due to the removal of an iteration process in the nonlinear scheme. The second one is the introduction of the adaptivity in the algorithm both temporally and spatially — i.e. an h-adaptive mesh method is employed to control the total amount of mesh grids, and an adaptive stop criterion in time propagation process is designed based on an observation that total energy always decays much faster at the beginning. Plenty of numerical experiments successfully demonstrate effectiveness of our method.

AMS subject classifications: 65N30, 37M05

Key words: Kohn-Sham density functional theory, gradient flow model, structure-preserving, linear scheme, adaptive strategy.

1. Introduction

The Kohn-Sham density functional theory has been widely used in electronic structure

*Corresponding author. *Email address:* zhouj@xtu.edu.cn (J. Zhou)

calculations [22]. Due to the nonlinearity of the governing equation and the complexity of electronic structure system, numerical solution became a main research area in the density functional theory, which plays an important role in applications such as the design of functional materials and new energy development [10, 12].

The solution of the Kohn-Sham equation [2, 27] by self-consistent field (SCF) iteration is a popular approach to determine the ground state of electronic structure systems. It has been realized in various mature software, including DFT-FE[†], VASP[‡], and Quantum Espresso [14]. Several successful techniques have been developed for accelerating the simulation, such as adaptive mesh methods [1, 3, 23, 24], density mixing [20] and quality solvers for generalized eigenvalue problems [8] with effective preconditioners [16] in SCF iteration, Chebyshev filtering [29]. In spite of its popularity, the orthogonality constraint in solving Kohn-Sham equation brings difficulty on fully taking advantage of hardware when designing parallel algorithms. Another widely used approach is the imaginary time propagation (ITP) method [18, 21]. The Wick rotation allows to transfer the complex-valued time-dependent Kohn-Sham equation into a real-valued one. The ground state of a given system can be obtained when the imaginary time approaches infinity.

Besides solving the Kohn-Sham equation, minimizing total energy of the system attracts more and more attention recently. Several pioneer works are based on this idea. For example, in [5, 28], Zhou group focused on gradient type methods and conjugate gradient method for electronic structure calculations. Yang *et al.* [26] developed a new direct constrained optimization algorithm based on the projection of the total energy functional into a sequence of subspaces and finding the minimizer of the energy functional on every subspace. A new iteration method based on the gradient on the Stiefel manifold has been proposed in [28]. The main advantage of this approach is that the method avoids solving nonlinear eigenvalue problems, so that the main cost is the assembling of the total energy functional and operations on a manifold. However, it should be noted that [28] requires an explicit operation in order to preserve the orthogonality of the wave functions and this diminishes the efficiency of the simulations. To overcome this bottleneck, an infeasible method was proposed for minimizing the total energy of a Kohn-Sham system [13], so that the ground state can be gradually obtained, without explicit treatment of the orthogonality.

It is mentioned that recently an extended gradient flow based model for electronic structure calculations was proposed in [6], which is a time evolution problem and different from either nonlinear eigenvalue problem or energy minimization problem. It is shown [6] that this extended gradient exponentially decays over time t, and the equilibrium point of this model corresponds to a local minimizer of the Kohn-Sham energy functional. Even better, it has shown that the orthogonality of those wave functions can be automatically preserved during the simulation. Furthermore, the gradient flow based model was used in a general framework of orthogonality preserving schemes for electronic structure calculations — cf. [7]. All these features demonstrate this method potential in large system simulations.

An implicit midpoint scheme employed in [6] for temporal discretization showed the effectiveness of the algorithm. However, the room for further improvements of the algo-

[†]`https://github.com/dftfeDevelopers/dftfe`
[‡]`http://cms.mpi.univie.ac.at/vasp`