Workshop on Mathematical and Numerical Analysis of Electronic Structure Models

Venue: Pingjiang Room, Ruiyun Building, Pingjiang Fu, Suzhou

June 10—15, 2019

Date	Time	Content
June 11 (Tue.)	**08:50－09:00**	**Opening Ceremony**
	09:00－12:00	**Chair: Yvon Maday**
	09:00－09:50	Paul W. Ayers: Flexible wavefunction ansatze for N-electron systems
	09:50－10:20	Tea Break
	10:20－11:10	Mathieu Lewin: The local density approximation in density functional theory
	11:10－12:00	Huajie Chen: Asymptotics of one-body reduced density matrices in density functional theory
	12:00－15:00	Lunch and Free Discussion: Xiangyuan Cafes, Western Restaurant
	15:00－18:00	**Chair:Ya-xiang Yuan**
	15:00－15:50	Xiaoying Dai: Adaptive step size strategies for line search methods and their applications to electronic structure calculations
	15:50－16:40	Xin Liu: Parallelizable second-order approach for optimization problems with orthogonality constraints
	16:40－17:10	Tea Break
	17:10－18:00	Martin Gander: On the scalability of one level domain decomposition methods
	18:20－20:00	Dinner: Xiangyuan Cafes, Western Restaurant

Date	Time	Content
June 12 （Wed.）	**09:00－12:00**	**Chair: Eric Cances**
	09:00－09:50	Paul Cazeaux: Reduced modeling for electronic conductivity calculations in 2D hetero structures
	09:50－10:20	Tea Break
	10:20－11:10	Yuzhi Zhou: Plane wave methods for quantum eigenvalue problem of incommensurate systems
	11:10－12:00	Daniel Massatt: Momentum space in relaxed incommensurate bilayers
	12:00－15:00	Lunch and Free Discussion: Xiangyuan Cafes, Western Restaurant
	15:00－18:00	**Chair: Chao Yang**
	15:00－15:50	Xingyu Gao: The adaptive self-consistent field iteration for solving the Kohn-Sham equation
	15:50－16:40	Antoine Levitt: Screening and charge sloshing in density functional theory
	16:40－17:10	Tea Break
	17:10－18:00	Geneviève Dusson: Data-driven interatomic potentials based on symmetric polynomials
	18:20－20:00	Dinner: Daru Room, Ruiyun Building

Date	Time	Content
June 13 （Thu.）	**09:00－12:00**	**Chair: Gero Friesecke**
	09:00－09:50	Zhaojun Bai: Progress and challenge in eigenvector-dependent eigenvalue problems
	09:50－10:20	Tea break
	10:20－11:10	Chao Yang: Eigenvalues of an infinite dimensional tensor eigenvalue problem
	11:10－12:00	Agnieszka Miedlar: A rational approximation method for large-scale nonlinear eigenvalue problems
	12:00－14:00	Lunch: Xiangyuan Cafes, Western Restaurant
	14:00－18:00	Free Discussion

Date	Time	Content
June 14 （Fri.）	**09:00－12:00**	**Chair: Reinhold Schneider**
	09:00－09:50	Jianfeng Lu: Coordinate descent full configuration interaction
	09:50－10:20	Tea break
	10:20－11:10	Lin Lin: Fast semi-local and hybrid functional real-time TDDFT with the parallel transport gauge formulation
	11:10－12:00	Jun Fang: Implementation of the projector Augmented-wave method: the use of atomic data sets in the standard PAW-XML
	12:00－15:00	Lunch and Free Discussion: Xiangyuan Cafes, Western Restaurant
	15:00－18:00	**Chair: Mathieu Lewin**
	15:00－15:50	Ivan Duechemin: Resolution of the identity: old ideas and new concepts
	15:50－16:40	Jack Robert Thomas: Relaxation of a crystalline defect in the tight binding model
	16:40－17:10	Tea break
	17:10－18:00	Andre Laesteadius: Local analysis of coupled-cluster methods
	18:20－20:00	Dinner: Xiangyuan Cafes, Western Restaurant

Date	Time	Content
June 15 （Sat.）	**09:00－12:00**	**Chair: Zhaojun Bai**
	09:00－09:50	Jingrun Chen: Multiscale finite element methods for the semi-classical Schrodinger equation
	09:50－10:20	Tea break
	10:20－11:10	Clotilde Fermanian Kammerer: Propagation of wave packets and application to Herman-Kluk representation of the Schrödinger propagator
	11:10－12:00	Zaiwen Wen: Some recent progress on optimization from electronic structure calculation
	12:00－14:00	Lunch: Xiangyuan Cafes, Western Restaurant
	18:20－20:00	Dinner

Flexible wavefunction ansatze for N-electron systems

Paul W. Ayers, Taewon David Kim, and Ramón Alan Miranda Quintana

Department of Chemistry & Chemical Biology, McMaster University

Abstract

A flexible ansatz for N-electron wave functions that subsumes the wave functions used in traditional configuration interaction approaches, coupled cluster methods, geminal-product wave functions, electron-group-function approaches, matrix-product states, and tensor-network states is presented. In this approach, desirable properties of wave functions (e.g., quasiparticle interpretations; size-consistency) are expressed as properties of a function that determines the coefficients of Slater determinants as (generally nonlinear) functions of the input parameters. Some of these key properties will be presented, allowing us to generate entirely new wave function forms that have desirable formal properties and yet, in some cases, are fully applicable to strongly correlated systems. A perturbative form of this approach will also be presented; the perturbative expression is especially useful for generating initial guesses for the wave function optimization.

Progress and challenge in eigenvector-dependent eigenvalue problems

Zhaojun Bai

University of California at Davis

Abstract

Nonlinear eigenvalue problems with eigenvector nonlinearity (NEPv) arise in electronic structure calculations and machine learning. The NEPv is a much less explored topic compared to nonlinear eigenvalue problems with eigenvalue nonlinearity. From a linear algebra point of view, we will start this talk with recent progress in NEPv, such as the existence and uniqueness, local and global convergence analysis of the self-consistent field iteration. Then we will discuss a number of practical issues and challenges for solving NEPv, such as parallel algorithms for extracting many eigenpairs and handling of ill-conditioning.

Reduced modeling for electronic conductivity calculations in 2D heterostructures

Paul Cazeaux

University of Kansas

Abstract

Weak van der Waals interactions between 2D materials do not impose limitations on integrating highly disparate atomically thin layers such as graphene, phosphorene, boron-nitride... This is both a blessing, allowing the realization of many stable assemblies, but also a computational curse due to the loss of periodicity.
We will discuss how the computation of relaxation patterns, parameterization of tight-binding electronic structure models, and abstract C*-algebra representations can be efficiently implemented within a unified framework. This allows us in particular to reduce the complexity of evaluating Kubo formulae to compute macroscopic observables such as conductivity.

Asymptotics of one-body reduced density matrices in density functional theory

Huajie Chen

Beijing Normal University

Abstract

In this talk, we investigate the one-body reduced density matrices in density functional theory. The exact kinetic energy of a many-electron system is determined by the one-body reduced density matrix, which is not well-approximated in standard Kohn–Sham density functional models for strongly correlated systems. We study the exact one-body reduced density matrices for one dimensional electron gas, and derive the asymptotic decay rate of the natural occupation spectrum for the one-dimensional finite wigner crystal. This is a joint work with Gero Friesecke in TU Munich.

Multiscale finite element methods for the semi-classical Schrodinger equation

Jingrun Chen

Soochow University

Abstract

In recent years, an increasing attention has been paid to quantum heterostructures with tailored functionalities, such as heterojunctions and quantum metamaterials, in which quantum dynamics of electrons can be described by the Schrodinger equation with multiscale potentials. The model, however, cannot be solved by asymptotic-based approaches where an additive form of different scales in the potential term is required to construct the prescribed approximate solutions. In this paper, we propose a multiscale finite element method to solve this problem in the semiclassical regime. The localized multiscale basis functions are constructed using sparse compression of the Hamiltonian operator, and thus are ``blind" to the specific form of the potential term. After an one-shot eigendecomposition, we solve the resulting system of ordinary differential equations explicitly for the time evolution. In our approach, the spatial mesh size is $ H=\mathcal{O}(\epsilon) $, where ϵ is the semiclassical parameter and the time stepsize $ k$ is independent of ϵ. Numerical examples in one dimension with a periodic potential, a multiplicative two-scale potential, and a layered potential, and in two dimension with an additive two-scale potential and a checkboard potential are tested to demonstrate the robustness and efficiency of the proposed method. Moreover, first-order and second-order rates of convergence are observed in H^1 and L^2 norms, respectively. This is joint work with Dingjiong Ma and Zhiwen Zhang.

Adaptive step size strategies for line search methods and their application on electronic structure calculations

Xiaoying Dai

Academy of Mathematics and Systems Science, CAS

Abstract

In this talk, we will introduce an adaptive step size strategy for a class of line search methods for orthogonality constrained minimization problems. Under some mild assumptions, we prove the convergence of the line search method with our adaptive step size strategy. We then apply this new algorithm to electronic structure calculations of some typical systems. The numerical results show the efficiency of this new algorithm and its advantage to the existing step size strategy. This is a joint work with Liwei Zhang and Aihui Zhou.

Resolution of the identity: old ideas and new concepts.

Ivan Duchemin

French Alternative Energies and Atomic Energy Commission in Grenoble

Abstract

The introduction of auxiliary bases to approximate molecular orbital products has paved the way to significant savings in the evaluation of four-center two-electron Coulomb integrals. We first present a generalized dual space strategy that sheds a new light on variants over the standard density and Coulomb-fitting schemes, including the possibility of introducing minimization constraints or hybrid scheme, and explore the merits of these approaches on the basis of extensive Hartree–Fock and MP2 calculations over a standard set of medium size molecules. We finally discuss the introduction of separable dual measures and their implication in the reduction of computational complexity of specific electronic quatities.

Data-driven interatomic potentials based on symmetric polynomials

Geneviève Dusson

University of Warwick

Abstract

For many applications ab initio computations are too expensive, so that interatomic potentials, often cheap to compute but also less accurate, are in use. Originally derived from empirical models, these potentials have recently mainly been developed from data-driven methods, in the aim of combining the high accuracy of ab initio methods with the low computational cost of interatomic potentials.

In this talk, I will present the construction of interatomic potentials for materials systems relying on 1-the choice of a functional form based on a body-order expansion satisfying the symmetries of the problem, namely the invariance with respect to the permutation and rotation invariance of the system, 2-a polynomial regression fitted to condensed phase ab-initio data. I will report some numerical tests on two different databases (composed of W and Si), illustrating the accuracy, the low computational cost, and the systematic improvability of the potential.

Implementation of the projector augmented-wave method: the use of atomic datasets in the standard PAW-XML format

Jun Fang

Institute of Applied Physics and Computational Mathematics

Abstract

The projector augmented-wave (PAW)method is an important approach for electronic structure calculations based on the Kohn – Sham density functional theory. And the PAW atomic dataset plays an essential role in the implementation and application of the method. The intensive use of proprietary datasets with limited metadata in previous years has led to difficulties in both the cross-validation of PAW codes and the understanding of the accuracy and transferability of PAW atomic data. In this work, we focus on the open-source ABINIT Jollet – Torrent – Holzwarth dataset (JTH) library in the PAW-XML format and investigate the implementation techniques to clarify how the atomic data participate in the computations. We propose an intermediate dataset that extends the original PAW-XML one by atomic quantities in derived forms, which facilitate the PAW implementation using the JTH library and cover the differences between PAW datasets. Our implementation is validated by comparing the structural property results of representative bulk materials and molecules with those calculated by ABINIT using the same datasets. Moreover, we discuss the feasibility of using the intermediate dataset for a quick support of PAW-XML datasets in existing PAW codes.

On the scalability of one level domain decomposition methods

Martin J. Gander

Universite de Geneve

Abstract

It is well accepted in the domain decomposition community that one level domain decomposition methods are not scalable: if one increases the number of subdomains, more iterations are needed for the domain decomposition method to converge. A recent observation by Cancès, Maday and Stamm using a one level Schwarz method for the simulation of solvation models showed however that in this case the one level method is scalable. Together with Ciaramella, we proved this surprising scalability using Fourier analysis, maximum principle techniques and also the method of projections. I will first recall these results, and then show that all other main one level domain decomposition methods, like Dirichlet-Neumann, Neumann-Neumann, FETI and optimized Schwarz are also scalable in this case. I will finally explain why there are other cases where one level domain decomposition methods can be scalable: scalability depends on the partial differential operator, the geometry, and the boundary conditions imposed on the problem.

The adaptive self-consistent field iteration for solving the Kohn-Sham equation

Xingyu Gao

Institute of Applied Physics and Computational Mathematics

Abstract

Solving the Kohn-Sham equation is most computationally demanding in the first-principles calculations. The self-consistent field (SCF) iteration is used to solve such a nonlinear eigenvalue problem. With some fundamental info on the system, we develop a modified Kerker preconditioning scheme which captures the long-range screening behavior of inhomogeneous systems and thus improves the SCF convergence. For situations without a priori knowledge of the system, we design the a posteriori indicator to monitor if the preconditioner has suppressed charge sloshing during the iterations. Based on the a posteriori indicator, we demonstrate two schemes of the self-adaptive configuration for the SCF iteration. This is a joint work with Yuzhi Zhou, Han Wang, Yu Liu and Haifeng Song.

Propagation of wave packets and application to Herman-Kluk representation of the Schrödinger propagator

Clotilde Fermanian Kammerer

Paris Est-Crteil University

Abstract

In this talk, we present recent results obtained in collaboration with Caroline Lasser (Technische Universität München) and Didier Robert (Université de Nantes). We shall describe (old and new) results about the propagation of coherent states through systems of Schrödinger equations, including situations with codimension 1 crossings. In the scalar case, it is well-known that such a description can be used for deriving integral representations of the Schrödinger propagator, the so-called Hermann-Kluk representations, that naturally adapts to numerical realisations. We shall explain how such an approach extends to the case of systems.

Local analysis of coupled-cluster methods

A. Laestadius^1, F. Faulstich1^, S. Kvaal^1, O. Legeza^2, R. Schneider^3

1 University of Oslo, Department of Chemistry

2 Wigner Research Center for Physics, Strongly Correlated Systems

3 Technische Universität Berlin, Department of Mathematics

Abstract

The coupled-cluster (CC) method and its variations have been analyzed [1-4] within the ERC project BIVAQUM [5]. This project studies a generalized variational principle, the so-called bivariational principle, where the bra and ket wave functions of the Rayleigh-Ritz quotient are treated as truly independent variables. We here aim at explaining the basic mathematical concepts used to prove a locally unique solution of different CC methods - including the extended CC and the tailored CC method. Such a local analysis addresses truncation schemes and their convergence. The key notion is local strong monotonicity. The connection to a HOMO-LUMO gap and the fluctuation potential, defined as the difference between the system's Hamiltonian and the Fock operator, is investigated.

References

[1] A. Laestadius and S. Kvaal, SIAM J. Numer. Anal. 56, 660, (2018).
[2] F.M. Faulstich, A. Laestadius, S. Kvaal, Ő. Legeza, and R. Schneider, arXiv:1802.05699, (2018).
[3] A. Laestadius and F.M. Faulstich, Molecular Physics (2019).
[4] F.M. Faulstich, M. Máté, A. Laestadius, M.A. Csirik, L. Veis, A. Antalik, J. Brabec, R. Schneider, J. Pittner, S. Kvaal, and Ő. Legeza, Journal of Chemical Theory and Computation (2019).
[5] S. Kvaal, http://www.bivaqum.no

Screening and charge sloshing in density functional theory

Antoine Levitt

Ecole des Ponts Paris Tech and INRIA Paris

Abstract

When a free positive charge is inserted in a metal, electrons flock towards it. This creates a reaction potential that effectively nullifies the Coulomb potential of the charge at long range. I will explain how this phenomenon occurs in the framework of the linear response of the reduced Hartree-Fock model (a simplified version of DFT) of defects at finite temperature. The analysis also sheds light on the convergence of algorithms to solve the self-consistent equations of DFT, and the phenomenon of "charge sloshing".

The local density approximation in density functional theory

Mathieu Lewin

University Paris Dauphine

Abstract

I will present the first mathematically rigorous justification of the Local Density Approximation (LDA) in Density Functional Theory. More precisely, I will discuss a quantitative bound on the difference between the Levy-Lieb functional and the LDA, involving gradient terms. The LDA becomes correct in the regime where the density is very flat on sufficiently large regions of space. Joint work with Elliott H. Lieb (Princeton) and Robert Seiringer (IST Vienna).

Fast semi-local and hybrid functional real-time TDDFT with the parallel transport gauge formulation

Lin Lin

University of California at Berkeley

Abstract

Real-time time-dependent density functional theory (rt-TDDFT) is known to be hindered by the very small time step (attosecond or smaller) needed in the numerical simulation, which significantly limits its range of applicability in the study of ultrafast dynamics. We demonstrate that RT-TDDFT calculations can be significantly accelerated using a combination of two techniques: parallel transport gauge and implicit time integrator, while neither technique would be sufficiently effective on its own. Using absorption spectrum and Ehrenfest dynamics calculations in a planewave basis set for example, we show that the new method can utilize a time step that is on the order of 10-100 attoseconds, and is no less than 5-10 times faster when compared to standard explicit time integrators. We also demonstrate that the parallel transport formulation enables large scale rt-TDDFT calculations with hybrid functional, which was previously considered to be prohibitively expensive. On the Summit supercomputer (No.1 supercomputer in the Top500 list in November 2018), we can carry out an rt-TDDFT+hybrid functional simulation with the planewave basis set for a unprecedentedly large system with 1536 silicon atoms, with a practical time to solution of 1.5 hours per femtosecond on 768 GPUs. (Joint work with Dong An, Weile Jia, Lin-Wang Wang)

Parallelizable second-order approach for optimization problems with orthogonality constraints

Xin Liu

Academy of Mathematics and Systems Science, CAS

Abstract

Updating the augmented Lagrangian multiplier by closed-form expression yields efficient infeasible approach for optimization problems with orthogonality constraints. Hence, parallelization becomes tractable in solving this type of problems. To accelerate the local convergence, we consider second-order approach under this framework. To avoid expensive calculation or solving a hard subproblem in computing the Newton step, we propose a new strategy to do it approximately which leads to superlinear convergence theoretically. In practice, the new second-order approach outperforms the existent algorithms. Last but not least, this new approach is completely orthonormalization-free and hence can be parallelized directly.

Coordinate descent full configuration interaction

Jianfeng Lu

Duke University

Abstract

The leading eigenvalue problems arise in many applications. When the dimension of the matrix is super huge, such as for applications in quantum many-body problems, conventional algorithms become impractical due to computational and memory complexity.
In this talk, we will describe some recent works on new algorithms for the leading eigenvalue problems based on randomized and coordinate-wise methods (joint work with Yingzhou Li and Zhe Wang).

Momentum space in relaxed incommensurate bilayers

Daniel Massatt

University of Chicago

Abstract

Momentum space techniques are fundamental to the understanding of electronic structure of materials. The basic formulation of momentum space comes from the Bloch operator for periodic materials. It has been shown momentum space can be formulated for homogenous incommensurate bilayers, and has even begun to be extended to homogenous trilayers, thus extending momentum space beyond the periodic regime. Here we go further by removing the requirement for homogeneity in the layers, extending momentum space to include mechanical relaxation effects in the incommensurate bilayer systems. We also fully formalize the momentum space technique, showing it is composed of a scattering description coming directly from the monolayer Bloch operators. This formalism can be easily extended to other problems with scattering such as phonons and multilayers.

A rational approximation method for large-scale nonlinear eigenvalue problems

Agnieszka Miedlar

University of Kansas

Abstract

Eigenvalue problems in which the coefficient matrices depend nonlinearly on the eigenvalues arise in a variety of applications in science and engineering, e.g., dynamic analysis of structures or computational nanoelectronics, to mention just a few.
This talk will discuss how the Cauchy integral-based approaches offer an attractive framework to develop highly efficient and flexible techniques for solving large-scale nonlinear eigenvalue problems. We will introduce the nonlinear counterpart of the well-established linear FEAST algorithm. Like its linear predecessor, the nonlinear FEAST (NLFEAST) algorithm can be used to solve nonlinear eigenvalue problems for the eigenpairs corresponding to eigenvalues that lie in a user-specified region in the complex plane, thereby allowing for the calculation of large number of eigenpairs in parallel. Finally, we will use several real-world examples to illustrate the method.
This is a joint work with B. Gavin, E. Polizzi and Y. Saad.

Relaxation of a crystalline defect in the tight binding model

Jack Robert Thomas

University of Warwick

Abstract

In (Chen, Lu, Ortner, 2016), a tight binding model for point defects is formulated in the canonical ensemble with finite temperature. The limiting model as domain size grows to infinity is shown to be formulated in the grand-canonical ensemble with Fermi level fixed corresponding to the perfect crystal. In the present work, we extend these results to the more common zero temperature models under the assumption that the Fermi level is not an eigenvalue of the limiting Hamiltonian. In particular, we formulate zero temperature models in the grand-canonical ensemble and prove these are consistent with taking zero temperature and thermodynamic limits in the finite temperature models. As an important by-product of the aforementioned results, we obtain qualitatively sharp estimates on the interaction range for the zero temperature electronic structure model.

Some recent progress on optimization from electronic structure calculation

Zaiwen Wen

Peking University

Abstract

In this talk, we will introduce: 1) efficient subspace extractions for matrix optimization problems with spectral or low-rank structures; 2) A Riemannian gradient method for finding the ground states and their characterization of spin-F Bose-Einstein condensates; 3) the geometric property of the minimization of the sum of a quadratic and quartic function over a single spherical constraint.

Eigenvalues of an infinite dimensional tensor eigenvalue problem

Chao Yang

Lawrence Berkeley National Laboratory

Abstract

We are interested in computing eigenvalues of an infinite dimensional matrix that can be written as an infinite sum of Kronecker products of infinite number of matrices. One way to solve such a problem is to use an infinite translational invariant tensor ring to represent the approximation to the eigenvector to be computed and update the core of such a tensor ring through a flexible power iteration. We discuss approximations made in each step of the power iteration and how these approximations affect the convergence of the method.

Plane wave methods for quantum eigenvalue problems of incommensurate systems

Yuzhi Zhou

CAEP Software Center for High Performance Numerical Simulation

Abstract

We propose a novel numerical algorithm for computing the electronic structure related eigenvalue problems of incommensurate systems. Unlike the conventional practice that approximates the system by a large commensurate supercell, our algorithm directly discretizes the eigenvalue problems under the framework of a plane wave method. The emerging ergodicity and the interpretation from higher dimensions give rise to many unique features compared to what we have been familiar with in the periodic systems. The numerical results of 1D and 2D quantum eigenvalue problems are presented to show the reliability and efficiency of our algorithm. Furthermore, the extension of our algorithm to full Kohn-Sham density functional theory calculations is discussed.