

MO 7: Machine Learning and Computational and Theoretical Molecular Physics

Time: Wednesday 11:00–13:00

Location: F142

Invited Talk

MO 7.1 Wed 11:00 F142

Augmenting basis with normalizing flows for solving Schrödinger equations: theoretical analysis — ●YAHYA SALEH^{1,2}, ARMIN ISKE², ANDREY YACHMENEV^{1,4}, and JOCHEN KÜPPER^{1,3,4} — ¹Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Mathematics, Universität Hamburg — ³Department of Physics, Universität Hamburg — ⁴for Ultrafast Imaging, Universität Hamburg

Spectral methods are a popular class for solving time-independent Schrödinger equations. Here, one approximates the wavefunctions in the linear span of standard basis sets of L2. In spite of the well-posedness and the convergence guarantees of such methods, they suffer from the curse of dimensionality, as the computational expenses grow exponentially with the size of the quantum system.

Recently, nonlinear functions, e.g., neural networks have been proposed to model ground states and low-lying excited states of Schrödinger equations. Although they promise accurate results with smaller scaling than standard methods, extensions of such models to the simultaneous computation of many states are still lacking.

Here, we propose to model excited states of Schrödinger equations via augmented basis sets, where standard basis sets are composed with normalizing flows. We show that such a numerical scheme is well-posed and defines a richer approximation space than standard methods. Moreover, we provide convergence guarantees.

MO 7.2 Wed 11:30 F142

A machine learning full dimensional potential energy surface for AlF-AlF: lifetime of the intermediate complex — WEIQI WANG¹, ●XIANGYUE LIU¹, and JESÚS PÉREZ-RÍOS² — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²Institute for Advanced Computational Science, Stony Brook University, Stony Brook, NY 11794-3800, USA

AlF is a promising candidate in the quest of finding the most efficient molecule for laser cooling. In this work, a full-dimensional potential energy surface of AlF-AlF dimer has been constructed by machine learning methods. In particular, we analyze the reliability and efficiency of different active learning schemes developed for this system. The potential energy surface has been employed in calculating the four-body complex lifetime relevant to the stability of molecules in the ultracold regime via molecular dynamics simulations.

MO 7.3 Wed 11:45 F142

Quantum flows neural network for variational solutions of the Schrödinger equation — ●ÁLVARO FERNÁNDEZ^{1,3}, YAHYA SALEH^{1,4}, ANDREY YACHMENEV^{1,2}, ARMIN ISKE⁴, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Center for Ultrafast Imaging, Universität Hamburg — ³Department of Physics, Universität Hamburg — ⁴Department of Mathematics, Universität Hamburg

Recently, a few deep neural network models for solving the electronic Schrödinger equation were developed, demonstrating both outstanding computing efficiency and accurate results.

Here, we present a new quantum-flow-neural-network approach for obtaining variational solutions of the stationary Schrödinger equation. At the core of the method is an invertible neural network composed with the general basis of orthogonal functions, which provides a more stable framework for simultaneous optimization of the ground state and excited states. This approach is applied in calculations of the vibrational energy levels of polyatomic molecules as well as of electronic energies in a single-active-electron approximation. The results show a considerable improvement of variational convergence for the ground and the excited states. In addition, we extend our approach for solving the time dependent problems using recurrent flows.

MO 7.4 Wed 12:00 F142

Electronic excited states in deep variational Monte Carlo — ●MIKE ENTWISTLE¹, ZENO SCHÄTZLE¹, PAOLO ERDMAN¹, JAN HERMANN^{2,1}, and FRANK NOÉ^{2,1,3} — ¹Freie Universität Berlin, Berlin, Germany — ²Microsoft Research AI4Science, Berlin, Germany — ³Rice University, Houston, USA

Obtaining accurate ground and low-lying excited states of electronic systems is crucial in a multitude of important applications. One ab ini-

tio method for solving the Schrödinger equation that scales favorably for large systems is variational quantum Monte Carlo (VMC). The recently introduced deep VMC approach uses ansatzes represented by deep neural networks and generates nearly exact ground-state solutions for molecules containing up to a few dozen electrons, with the potential to scale to much larger systems where other highly accurate methods are not feasible. Here, we extend one such ansatz (PauliNet) to compute electronic excited states. We demonstrate our method on various small atoms and molecules and consistently achieve high accuracy for low-lying states. To highlight the potential of our method, we compute the first excited state of the much larger benzene molecule, as well as the conical intersection of ethylene, with PauliNet matching results of more expensive high-level methods.

MO 7.5 Wed 12:15 F142

The performance of CCSD(T) for the calculation of dipole moments in diatomics — ●XIANGYUE LIU¹, LAURA MCKEMMISH², and JESÚS PÉREZ-RÍOS³ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²School of Chemistry, UNSW Sydney, Sydney, NSW 2052, Australia — ³Department of Physics and Astronomy, Stony Brook University, Stony Brook 11794, New York, USA

Electric dipole moment plays an important role in understanding intermolecular interactions. High-quality electric dipole moments are essential for accurate predictions of vibrational and rotational spectroscopy. In this work, the performance of CCSD(T) (coupled cluster with single, double, and perturbative triple excitations) has been evaluated with accurate experimental data of diatomic molecules. In particular, CCSD(T) accuracy for the equilibrium bond length, vibrational frequency, and dipole moments has been discussed. We find that CCSD(T) gives accurate predictions on dipole moments for most of the molecules in the test set. However, disagreements have been found for a few molecules, which can hardly be explained by relativistic or multi-reference effects. The impacts of basis set family and size have also been discussed.

MO 7.6 Wed 12:30 F142

Non-Local Polarizability Density as a Building Block for Dispersion Density Functionals — ●SZABOLCS GÖGER¹, PETER SZABO^{2,3}, DMITRY FEDOROV¹, and ALEXANDRE TKATCHENKO¹ — ¹University of Luxembourg, 1511 Luxembourg, Luxembourg — ²Katholieke Universiteit Leuven, 3000 Leuven, Belgium — ³Royal Belgian Institute for Space Aeronomy, 1180 Uccle, Belgium

Dispersion interactions, stemming from long-range electron correlations, are not properly captured by many electronic structure methods. A proper framework to tackle this problem requires determining the correlation energy via the adiabatic-connection dissipation-fluctuation theorem, but a robust unified formalism is yet to be developed [1]. In this work, we attempt to build such a general method based on the non-local polarizability, which is expressed as a polarization-polarization correlation function. This quantity is evaluated for a number of model systems and contrasted to real atoms and molecules. The model studies presented here, along with prior work on semi-local polarizability functionals [2], pave the way toward developing a unified non-local polarizability functional for molecules and materials, aimed to describe the short-range and long-range correlations on equal footing.

[1] Hermann and Tkatchenko, Phys. Rev. Lett. 124, 146401 (2020)

[2] Vydrov and Van Voorhis, Phys. Rev. Lett. 103, 063004 (2009)

MO 7.7 Wed 12:45 F142

Few-Body Physics of the Trapped Atoms: The Configuration Interaction Approach — ●MATEE UR REHMAN¹, ALEJANDRO SAENZ¹, FABIO REVUELTA PEÑA², PAUL WINTER¹, and SIMON SALA¹ — ¹Humboldt-Universität zu Berlin — ²Universidad Politécnica de Madrid

Two independent theoretical approaches are developed for the computational treatment of interacting ultracold atoms in versatile trap potentials. One approach considers the two-body system in centroid and relative coordinates have recently successfully demonstrated the inelastic confinement-induced resonances occurs due to the anharmonicity of the trap potentials, however an extension beyond two particles is not possible, but evidently of interest. This motivates to consider an alter-

native approach in absolute coordinates based on standard quantum-chemistry approaches, that uses cartesian Gaussians (GTOs) as basis functions and performs the configuration interaction calculations with a flexible choice of interaction potentials, hence allow considering arbitrary optical tweezer arrays. As the delta-pseudopotential does not work in beyond-mean field description, the Gaussian interaction potential using GTOs allows an efficient solution of multi-centered

six-dimensional interaction integrals, but its validity has to be investigated. So this talk will demonstrate both theoretical models by focusing on their pros and cons for different interatomic interaction potentials, and then address the question that, Is the Gaussian interaction potential is sufficient (respectively in which parameter regime is it sufficient) to be used (or within which accuracy it can be used).