CPP 74: Focus Session: Frontiers of Electronic-Structure Theory: Correlated Electron Materials VII (joint session O/TT/MM/DS/CPP)

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(Synopsis provided with part I of this session)

Time: Thursday 15:00–17:45 Location: HL 001

Invited Talk CPP 74.1 Thu 15:00 HL 001

Recent developments in FCIQMC: real-time propagation and improved convergence with walker number — •Ali Alavi — Max Planck Institute for Solid State Research, Stuttgart, Germany

The Full Configuration Interaction QMC method samples Slater determinants using an imaginary-time propagation of walkers, and can yield essentially exact ground- and excited states energies and wavefunctions for Fermionic systems. Recently we have extended this methodology to real-time propagation, enabling the calculation of spectral functions along the real-frequency axis. This method will be described in the talk, together with representative examples from molecular and lattice models. We will also describe a second development in the FCIQMC methodology which substantially improves the rate of convergence of the ground-state technique with respect to the number of walkers. With the new method, we can compute essentially the exact ground state energy of the benzene molecule, correlating 30 electrons (the entire valence) in the full set of 108 orbitals of a VDZ basis. Perspectives of the new methods will be discussed.

CPP 74.2 Thu 15:30 HL 001

Quasi-Continuous LDA+DMFT calculations for SrVO3. — •Evan Sheridan, Christopher Rhodes, Evgeny Plekhanov, and Cedric Weber — King's College London, Theory and Simulation of Condensed Matter (TSCM), The Strand, London, United Kingdom.

The Dynamical Mean Field Theory (DMFT) is an extremely powerful tool in the treatment of strongly correlated electron systems and many DMFT calculations suffer from a computational bottleneck when it attempts to solve the Anderson Impurity Model (AIM).

Common among the early Anderson Impurity solvers was the Auxiliary Field Quantum Monte Carlo (AF-QMC) approach which relies on a discretisation of the imaginary time grid. AF-QMC solvers suffer from the notorious Suzuki-Trotter error,as a result of this , that has largely been ignored in recent years with the advent of Continuous Time-QMC (CT-QMC) solvers.

Here, we present a systematic study of how this issue can be overcome for realistic material properties using LDA+DMFT. We find that our quasi-continuous time method compares well to the state-of-the-art CT-QMC calculations for SrVO3, with the added advantage of linear scaling in temperature. The theoretical framework proposed is quite general and can be extended to cluster DMFT calculations.

CPP 74.3 Thu 15:45 HL 001

High temperature superconducting oxychlorides: a light element model for cuprates — •Matteo d'Astuto[1,2], Blair Lebert[2,3], Ikuya Yamada[4], and Masaki Azuma[5] — [1]Institut NEEL CNRS/UGA UPR2940 25 rue des Martyrs BP 166 38042 Grenoble cedex 9 FRANCE — [2]IMPMC, UMR7590 UPMC-Sorbonne Universités - CNRS, Paris, France — [3]Synchrotron SOLEIL, Gif-sur-Yvette, France — [4]Nanoscience and Nanotechnology Research Center (N2RC), Osaka, Japan — [5]Materials and Structures Laboratory, TITech, Yokohama, Japan

The copper oxychloride cuprate $Ca_2CuO_2Cl_2$ (CCOC) system, with vacancy or Na doping on the Ca site, is unique among the high temperature superconducting cuprates (HTSCs) since it: lacks high Z atoms; has a simple I4/mmm 1-layer structure, typical of 214 (LSCO) cuprates, but which is stable at all doping and temperatures; and has a strong 2D character due to the replacement of apical oxygen with chlorine. It also shows a remarkable phase digram, with a superconducting T_C growing to the optimal doping without any minimum around 1/8 doping, despite the observation of charge modulations by near-field spectro-microscopy. Due to the reduced number of electrons, advanced calculations that incorporate correlation effects, such as quantum Monte Carlo are easier, but relatively little is known about CCOC (for a cuprate) from an experimental point of view. We are now filling this gap by a comprehensive experimental study covering the whole phase diagram, in particular of the (para)magnon and phonon dispersion..

CPP 74.4 Thu 16:00 HL 001

Antiferromagnetic correlations in the metallic strongly correlated transition metal oxide $LaNiO_3$ — •Hanjie Guo[1], Zhiwei Li[1], Li Zhao[1], Zhiwei Hu[1], Chunfu Chang[1], Changyang Kuo[1], Wolfgang Schmidt[2], Andrea Piovano[2], Tunwen Pi[3], Oleg Sobolev[4], Daniel Khomskii[1], Liu Hao Tjeng[1], and Alexander Komarek[1] — [1]MPI CPfS, Dresden, Germany — [2]ILL, Grenoble, France — [3]NSRRC, Taiwan — [4]FRMII, Munich, Germany

The material class of rare earth nickelates with high Ni^{3+} oxidation state is generating continued interest due to the occurrence of a metal-insulator transition with charge order and the appearance of non-collinear magnetic phases within this insulating regime. The recent theoretical prediction for superconductivity in $LaNiO_3$ thin films has also triggered intensive research efforts. $LaNiO_3$ seems to be the only rare earth nickelate that stays metallic and paramagnetic down to lowest temperatures. So far, centimetre-sized impurity-free single crystal growth has not been reported for the rare earth nickelates material class since elevated oxygen pressures are required for their synthesis. Here, we report on the successful growth of centimetre-sized $LaNiO_3$ single crystals by the floating zone technique at oxygen pressures of up to 150 bar. Our crystals are essentially free from Ni^{2+} impurities and exhibit metallic properties together with an unexpected but clear antiferromagnetic transition.

CPP 74.5 Thu 16:15 HL 001

First-principles quantum Monte Carlo study of correlated materials — •Huihuo Zheng — Argonne Leadership Computing Facility, Argonne National Laboratory, Lemont, USA

Strongly correlated electronic systems have become an important subject of condensed matter physics, because of many fascinating phenomena arising in these systems such as metal-insulator transition, high temperature superconductivity, etc. Accurate characterization of the electron-electron correlations in these systems from first principles is essential for us to understand how these phenomena emerge from microscopic interactions. I will present our efforts in modeling correlated materials using the first-principles quantum Monte Carlo (QMC) method by showing two representative *ab intio* studies (vanadium dioxide and graphene) and a density-matrix downfolding theory for constructing low energy effective models from *ab initio* simulations. Using QMC, we correctly characterized the electronic structure of vanadium dioxide and unveiled the electronic origin of the metal-insulator transition which has been a mystery for decades. For graphene, we computed the electron screening from σ bonding electrons and illustrated how the emergent physics from underlying Coulomb interactions results in the observed weakly correlated semimetal. On the other hand, the downfolding approach we developed provides a way to quantitatively identify important microscopic interactions relevant to the macroscopic physics.

CPP 74.6 Thu 16:30 HL 001

Reduced Density Matrix Theory for Coupled Fermion-Boson Systems — •Florian Buchholz[1], Iris Theophilou[1], Michael Ruggenthaler[1], Heiko Appel[1], and Angel Rubio[1,2,3] — [1]MPSD, Hamburg, Germany — [2]CCQ, The Flatiron Institute, New York, United Sates — [3]Nano-bio Spectroscopy Group, San Sebastián, Spain

Reduced density matrix (RDM) theory proved to be successful in describing a wide range of many-body problems that are not easily accessible by the more common many-body perturbation theories or density functional theory. Especially as RDM theories are non-perturbative, they are advantageous in strong coupling scenarios.

However, RDM theory was to our knowledge never applied to systems with more than one active particle type. The focus of this talk is to analyze the possibilities and problems of an extension to coupled fermion-boson theories. Comparing a typical bilinear interaction term

of the form $c_i^+ c_j(a_k^+ + a_k)$, where c^+/c and a^+/a indicate fermion and boson creation/annihilation operators, respectively and the fermionic 2-body interaction term $c_i^+ c_j^+ c_k c_l$, the former should have a considerably reduced definition space, which we hope to be exploitable. On the other hand, the bilinear interaction has a very different structure than the 2-body interaction and it is not clear at all, how to define a RDM that carries all information to compute experimental observables of a coupled fermion-boson system.

Specifically, I will illustrate some of the peculiarities of the fermion-boson interaction for simple model systems and present some ideas to deal with those.

CPP 74.7 Thu 16:45 HL 001

Critical temperatures as function of magnetic anisotropy in two-dimensional systems from first-principles calculations — •Daniele Torelli — Center for Atomic-Scale Materials Design, Department of Physics, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

Recent observation of ferromagnetic out-of-plane order in monolayer CrI_3 highlights the importance of a microscopic understanding of anisotropy in ground state magnetic systems. Single-ion anisotropy accounts mainly for spin-orbit coupling interaction and, in particular for two-dimensional (2D) materials, it's crucial to escape the Mermin-Wagner theorem. Here we investigate the variation of critical temperatures as functions of anisotropy in Heisenberg model systems using Metropolis Monte Carlo simulations. Results for square, hexagonal and honeycomb lattices are compared with equivalent simulations in the Ising model, which is confirmed to represent the limit with infinite anisotropy. Based on a new developed computational 2D materials database, we predict a vast number of 2D structures with high critical temperatures. As testing system, relevant Heisenberg exchange couplings and magnetic anisotropy energy in CrI_3 monolayer are extracted from first principle calculations and energy mapping analysis, yielding to an estimation of Curie temperature in good agreement with experimental results.

CPP 74.8 Thu 17:00 HL 001

Oxygen vacancy-induced absorption of visible light in SrNbO3 — •Marcello Turtulici, Steffen Backes, and Silke Biermann — Centre de Physique Théorique, Ecole Polytechnique, 91128 Palaiseau, France

SrNbO3 has recently attracted attention as a bright red photocatalyst. Several, mutually contradicting, models have been proposed in the literature in order to explain the strong absorption in the visible spectrum, and no consensus even on the basic nature of the mechanism has been reached. In this work we investigate the optical properties of this material by means of state-of-the-art Density Functional Theory and many-body perturbation theory techniques. We evidence a high sensitivity of the optical properties on deviations from the ideal crystal structure. In particular, the optical properties should strongly depend on the presence of oxygen vacancies, which give rise to additional absorption channels in the visible frequency range. Most notably, the experimentally observed red color is likely due to transitions between orbitals of dominant Nb-eg character, which are enhanced by the strong hybridization of the quite extended 4d-states of Nb with oxygen p-states.

CPP 74.9 Thu 17:15 HL 001

Transient charge and energy flow in the wide-band limit — Fabio Covito, •Florian Eich, Riku Tuovinen, Michael Sentef, and Angel Rubio — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Thanks to recent advances in ultra-fast pump-probe spectroscopies and nano-thermometry it is possible to study charge and energy flow at atomic time and length scales. In order to analyze the transient dynamics of nanoscale devices theoretically, the wide-band limit is a commonly used approximation. Here we investigate the applicability of the wide-band limit to the study of charge and heat transport through nanojunctions exposed to voltage biases and temperature gradients. We find that while this approximation faithfully describes the long-time steady-state charge and heat transport, it fails to characterize the short-time transient behavior of the junction. In particular, we find that the charge current flowing through the device shows a discontinuity when a temperature gradient is applied, while the energy flow is discontinuous when a voltage bias drives the dynamics and even diverges when the junction is exposed to both a temperature gradient and a voltage bias. We discuss this pathological behavior and propose two possible solutions.

CPP 74.10 Thu 17:30 HL 001

From DFT to Coupled Cluster Theory - Understanding Oxygen Activation on Coin Metal Nanoparticles — •Wilke Dononelli and Thorsten Klüner — Institut für Chemie, Carl von Ossietzky Universität Oldenburg, 26111 Oldenburg, Germany

In this study we focus on one of the most fundamental catalytic model reactions, the oxidation of CO on a metal catalyst. We studied the activation of molecular oxygen via dissociation or direct reaction of CO and O_2 within density functional theory (DFT) and high level CCSD(T) calculations. Therefore we use Au_{13} and Au_{55} nanoparticles (NPs) and a periodic Au(321) surface as model systems and compare the catalytic activity of the gold substrates to Ag and Cu based, as well as bimetallic NP catalysts. Part of the DFT calculations were performed, using the well-established PBE functional as implemented in the Vienna ab initio simulation package (VASP). Hybrid and double hybrid DFT calculations on the NPs were performed in Gaussian09. CCSD(T) calculation were performed in Gaussian09 using conventional CCSD(T) for the M_{13} (M=Au,Ag,Cu) NPs and CCSD(T)/PBE in a QM/QM embedding scheme using the ONIOM approach for M_{55} NPs. For systems of 55 metal atoms PBE gives the same results as double hybrids or even CCSD(T). For smaller M_{13} NPs interaction energies differ between PBE and higher levels of theory, which might be explained by the molecule like character of these NPs.