O 35: Focus Session: Frontiers of Electronic-Structure Theory III (joint session O/HL)

Time: Tuesday 10:30–12:45   Location: TRE Ma

O 35.1   Tue 10:30   TRE Ma
Two-component GW implementation for molecular valence excitations — ●Qinglong Liu, Ramón L. Panadés-Barrueta, and Dorothea Golze — Chair of Theoretical Chemistry, Technische Universit at Dresden, 01062 Dresden, Germany

We present an all-electron GW implementation for the computation of charged molecular excitations, that includes scalar relativistic effects and spin-orbit coupling (SOC). Our method is based on a two-component (2c) approach, which can process 2c spinors and their corresponding eigenvalues from different levels of theory. The relativistic input for our 2c GW calculation is obtained in two ways: One approach is a non-self-consistent second variation SOC scheme, i.e. a scalar relativistic (SR) calculation is performed followed by an expansion of the spinors on top of the SR eigenvectors [1]. Another approach is the spinors and their eigenvalues are obtained by running a self-consistent relativistic DFT calculation with the X2C method [2]. Our algorithm has been implemented in the FHI-aims program package, which is based on numeric atom-centered orbitals (NAOs). In our 2c GW algorithm we combine the all-electron NAO scheme with the resolution of the identity technique based on the Coulomb metric (RI-V) and use the analytical continuation to evaluate the G_0W_0 self energy. We present results for the numerical validation of our implementation and for the influence of the relativistic input (second variation SOC vs X2C) on the valence excitations of small heavy molecules.
[1] W. Huhn and V. Blum, Phys. Rev. Materials 1, 033803 (2017)
[2] M. Iliaš and T. Saue, J. Chem. Phys. 126, 064102 (2007)

O 35.2   Tue 10:45   TRE Ma
Screened potential in two-dimensional GW calculations within the LAPW framework — ●Ben Alex, Sven Lubeck, and Claudia Draxl — Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

To calculate two-dimensional (2D) materials in a three-dimensional periodic code, one has to consider periodically repeated layers. The calculation of quasiparticle energies for 2D materials within the GW approximation requires the introduction of a 2D cutoff for the Coulomb potential as the layers would otherwise interact with each other. This cutoff leads to a stronger q dependence of the dielectric function around $q = 0$ which requires much denser sampling of the first Brillouin zone. In order to address this issue, an analytic expression for the dielectric function was previously derived for a plane-wave basis [1]. This expression is then integrated numerically in a small region around $q = 0$. The goal of this work is to adapt this technique to the linearized augmented planewave + local-orbital (LAPW+lo) basis as implemented in the full-potential all-electron code **exciting**. We show that also in our case, we obtain a significant computational speedup. Furthermore, this approach is compared with an approach where the dielectric function is interpolated to a denser q-grid.
[1] F. A. Rasmussen et al., Phys Rev B 94, 155406 (2016).

O 35.3   Tue 11:00   TRE Ma
GW multipole approach for the frequency description of the dielectric screening — ●Claudia Cardoso[1], Dario A. Leon[2], Andrea Ferretti[1], Daniele Varsano[1], and Elisa Molinari[1] — [1]S3 Centre, Istituto Nanoscienze, CNR, 41125, Modena (Italy) — [2]Department of Mechanical Engineering and Technology Management, Norwegian University of Life Sciences, 1430, Ås (Norway)

In the present work, we discuss a numerical approach for GW calculations that takes into account the frequency dependence of the screening via a multi-pole approximation (MPA), an accurate and efficient alternative to current full-frequency methods, that overcomes several limitations of the plasmon pole approximation (PPA).
MPA was recently developed and validated for semiconductors[1]. We now extend the use of MPA to metallic systems by optimizing the frequency sampling for these class of materials and propose a simple method to include the zero q limit of the intra-band contributions. The good agreement between MPA and full frequency results for the calculations of quasi-particle energies, polarizability, self-energy and spectral functions in different metallic systems confirms the accuracy and computational efficiency of the method. Finally, we discuss the physical interpretation of the MPA poles through a comparison with experimental electron energy loss spectra for Cu.

[1] D. A. Leon, C. Cardoso, T. Chiarotti, D. Varsano, E. Molinari, A. Ferretti, Phys. Rev. B 104, 115157

Topical Talk   O 35.4   Tue 11:15   TRE Ma
Towards low-scaling GW calculations for 2D materials — ●Jan Wilhelm — Institute of Theoretical Physics, University of Regensburg

Semiconducting two-dimensional materials are an ideal platform to study excitons thanks to the strong exciton binding energy and good experimental accessibility of the excitons. The GW+Bethe-Salpeter approach (GW+BSE) has been successful in analyzing excitons in single-layer 2D materials [1], but the application of GW+BSE is challenging for 2D double layers and moiré structures [2]. This is because the large unit cells in these structures contain hundreds to thousands of atoms, resulting in a high computational cost for GW+BSE calculations. In this talk, I will present a low-scaling GW algorithm for 2D materials that potentially allows for the inclusion of more than a thousand atoms in the simulation [3]. This algorithm is based on localized basis functions and can handle periodic boundary conditions and the divergence of Coulomb interactions in the Brillouin zone. I will present first benchmark calculations.
[1] D. Y. Qiu, F. H. da Jornada, S. G. Louie, PRL 111, 216805 (2013). [2] Nat. Phys. 17, 720 (2021), Nature 608, 499 (2022), Science 376, 406 (2022), Nature 603, 247 (2022). [3] J. Wilhelm, D. Golze, L. Talirz, J. Hutter, C. Pignedoli, JPCL 9, 306 (2018), J. Wilhelm, P. Seewald, D. Golze, JCTC 17, 1662 (2021).

15 min. break

O 35.5   Tue 12:00   TRE Ma
Accelerating core-level GW calculations by combining the contour deformation with the analytic continuation of W — ●Ramón L. Panadés Barrueta and Dorothea Golze — Theoretische Chemie, Technische Universität Dresden, Bergstr. 66c, 01062 Dresden, Deutschland

Many-body methods, like the GW approximation, have recently proven to be a highly effective tool for computing core-level excitations [1]. In particular, the contour deformation (CD) is an efficient, scalable and numerically stable approach that has enabled core-level calculations on systems up to 100 atoms [2]. In this work, we reduce the scaling of CD applied to core-levels from $O(N^5)$ to $O(N^4)$, using an analytic continuation of the screened Coulomb interaction W [3]. The new method (CD-WAC) has been implemented in FHI-aims. CD-WAC has been extensively tested on well established benchmark sets like the GW100 and the CORE65, reporting MAEs of less than 5 meV with respect to CD. The theoretical scaling has been confirmed by performing scaling experiments on large acene chains and amorphous carbon. Speedups of 5 times have been attained with CD-WAC for the largest systems.
[1] D. Golze, M. Dvorak, and P. Rinke. Front. Chem., 7:377, 2019.
[2] D. Golze, J. Wilhelm, M.J. Van Setten, and P. Rinke. J. Chem. Theory Comput., 14(9):4856-4869, 2018.
[3] I. Duchemin and X. Blase. J. Chem. Theory Comput., 16(3):1742-1756, 2020.

O 35.6   Tue 12:15   TRE Ma
Many-Body Effects of Metals Investigated by Means of the GW Method — ●Zimo Zhou, Nakib Protik, and Claudia Draxl — Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

Even if semilocal exchange-correlation functionals of density-functional theory (DFT) can capture the overall band structure of many metals well, they fail to obtain the correct position of the d-bands. This leads, for instance, to the underestimation of the interband absorption onset in the corresponding optical spectra as shown for a set of elemental metals [1]. In this work, we provide a systematic investigation of the quasi-particle band structure and the optical properties of this set of materials. To this extent, the self-energy corrections to the DFT results are computed by the GW approach of many-body perturbation theory as implemented in the full-potential all-electron code exciting [2,3]. We show that the optical absorption spectra based on these quasi-particle bands remedy the shortcomings of semi-local DFT, accurately reproducing the experimental counterparts.
[1] W. S. M. Werner, K. Glantschnig, and C. Ambrosch-Draxl, J.

Phys. Chem. Ref. Data 38, 1013 (2009). [2] A. Gulans, S. Kontur, C. Meisenbichler, D. Nabok, P. Pavone, S. Rigamonti, S. Sagmeister, U. Werner, and C. Draxl, J. Phys: Condens. Matter 26, 363202 (2014). [3] D. Nabok, A. Gulans, and C. Draxl, Phys. Rev. B 94, 035418 (2016).

O 35.7   Tue 12:30   TRE Ma

Separable Resolution-of-Identity in an all-electron numeric atom-centered basis set framework — •Francisco Delesma[1], Dorothea Golze[2], and Patrick Rinke[1] — [1]Department of Applied Physics, Aalto University, Espoo, Finland — [2]Faculty of Chemistry and Food Chemistry, Technische Universität Dresden, Dresden, Germany

The resolution-of-identity (RI) is a common way in quantum chemistry and computational materials science to reduce the computational cost of two-electron Coulomb integrals, another central entity in computational quantum mechanics. In 2019, Duchemin and Blase proposed the separable-RI approach [1], which preserves the accuracy of the standard, global RI method with the Coulomb metric (RI-V) and permits the formulation of cubic-scaling random-phase approximation (RPA) and Green's function based GW approaches.

In this work, we present the first implementation of the separable-RI in an all-electron numeric atom-centered orbital framework. Separable-RI is implemented in the FHI-aims code [2] and optimized for massively parallel execution. We extend the separable-RI framework beyond the original Hartree-Fock (HF) and GW implementations of Duchemin and Blase to MP2 and RPA, SOSEX and CCSD. Our separable-RI total energies and GW quasiparticle energies for the Thiel test set of small organic molecules reproduce the exact two-electron Coulomb integral calculations within 1 meV or better.

[1] I. Duchemin and X. Blase, J. Chem. Phys. 150, 174120 (2019)
[2] V. Blum, et al, Comput. Phys. Commun. 180, 2175, (2009)