

Computer Programs in Physics



Green/WeakCoupling: Implementation of fully self-consistent finite-temperature many-body perturbation theory for molecules and solids[☆]

Sergei Isakov^{a,*,1}, Chia-Nan Yeh^c, Pavel Pokhilko^b, Yang Yu^a, Lei Zhang^a, Gaurav Harsha^b, Vibin Abraham^b, Ming Wen^b, Munkhorgil Wang^b, Jacob Adamski^b, Tianran Chen^d, Emanuel Gull^a, Dominika Zgid^{a,b}

^a Department of Physics, University of Michigan, Ann Arbor, MI 48109, USA

^b Department of Chemistry, University of Michigan, Ann Arbor, MI 48109, USA

^c Center for Computational Quantum Physics, Flatiron Institute, 162 5th Avenue, New York, NY 10010, USA

^d Department of Physics and Engineering, West Chester University, West Chester, PA 19383, USA

ABSTRACT

The accurate ab-initio simulation of molecules and periodic solids with diagrammatic perturbation theory is an important task in quantum chemistry, condensed matter physics, and materials science. In this article, we present the `WeakCoupling` module of the open-source software package `Green`, which implements fully self-consistent diagrammatic weak coupling simulations, capable of dealing with real materials in the finite-temperature formalism. The code is licensed under the permissive MIT license. We provide self-consistent GW (scGW) and self-consistent second-order Green's function perturbation theory (GF2) solvers, analysis tools, and post-processing methods. This paper summarizes the theoretical methods implemented and provides background, tutorials and practical instructions for running simulations.

Program summary

Program Title: `Green/WeakCoupling`

CPC Library link to program files: <https://doi.org/10.17632/2ysyhzww6t.1>

Developer's repository link: <https://github.com/Green-Phys/green-mbpt>

Programming language: C++17, CUDA, Python

Licensing provisions: MIT License

External routines/libraries: MPI >= 3.0, BLAS, Eigen >= 3.4.0, cmake >= 3.18, cuBLAS.

Nature of problem: The simulation of periodic solids and molecules using diagrammatic perturbation theory

Solution method: We present an open-source implementation of the fully self-consistent finite-temperature many-body perturbation theory formalism at the GW and second-order perturbation theory level.

1. Introduction

The accurate, ab-initio, and systematically improvable numerical simulation of realistic interacting quantum systems using methods beyond density functional theory (DFT) [1,2] is an active, challenging area of research in condensed matter physics, quantum chemistry, and materials science. These simulations offer a route to material-specific, predictive modeling of experimentally complex systems, where an understanding of both physics and chemistry is necessary for gaining fundamental insights and optimizing technological advances.

[☆] The review of this paper was arranged by Prof. Weigel Martin.

* Corresponding author.

E-mail addresses: siskakov@umich.edu (S. Isakov), cye@flatironinstitute.org (C.-N. Yeh), pokhilko@umich.edu (P. Pokhilko), umyangyu@umich.edu (Y. Yu), lzphy@umich.edu (L. Zhang), gharsha@umich.edu (G. Harsha), avibin@umich.edu (V. Abraham), wenn@umich.edu (M. Wen), munkhw@umich.edu (M. Wang), adamskij@umich.edu (J. Adamski), tchen@wcupa.edu (T. Chen), egull@umich.edu (E. Gull), zgid@umich.edu (D. Zgid).

¹ Current address: Department of Physics, University of Michigan, Ann Arbor, Michigan 48109, USA.

Diagrammatic perturbation theory [3–7] provides a rigorous theoretical framework for these simulations. The central object of this theory, the one-body Green's function, allows for the evaluation of multiple important quantities, such as total energies [8], one-body observables, and spectral information such as band structures and band gaps that relate the simulation results to experiments such as angle resolved photoelectron spectroscopy (ARPES) [9]. The evaluation of two-body observables, other than the total energy, requires either solving the Bethe-Salpeter equation [10–12] to obtain the two particle Green's function or applying the thermodynamic Hellmann–Feynman theorem [13]. The computational access to experimentally relevant observables makes Green's function methods valuable for a broad range of applications.

Self-consistent diagrammatic theories, in particular Φ - [14] or Ψ - [15] derivable methods, are particularly useful in this context since they guarantee thermodynamic consistency, conservation laws [16], and – in the absence of multiple stationary points in the Φ - or Ψ -functional – starting point independence achieved at self-consistency [17].

Traditionally the domain of applicability of Green's function methods was limited to weak interactions or high temperatures, where low-order perturbative diagrammatic expansions of the self-energy were shown to provide accurate results. Green's function methods, such as G_0W_0 , when applied to semiconductors yielded band gap values in much better agreement with experiment than the local density approximation (LDA), resulting in popularization of various approximations to the GW method in materials science [18,19].

In recent years, the domain of applicability of Green's functions was extended to include moderately and strongly correlated systems, due to the introduction of Green's function embedding approaches [20,21]. In these methods the low order perturbative expansion is performed for orbitals containing itinerant electrons, where correlations are weak, while non-perturbative corrections are introduced for a handful of orbitals containing localized electrons displaying strong correlations. Embedding methods such as the local density approximation with dynamical mean-field theory (LDA+DMFT) [22] proved successful in qualitatively describing metal-to-insulator transition (MIT) in many strongly correlated perovskites.

The framework of many-body perturbation theory was developed already in the 1950s and 1960s [23,14,24] and its applications to molecules [25] and solids started in the late 1960s [26]. However, the methods employed, such as GW [24,27–29], frequently contained severe approximations when applied to realistic systems or were applied without significant approximations but only to model Hamiltonians. Among the most common approximations employed were: performing only the first iteration of GW, thus truncating the bold diagrammatic series at G_0W_0 ; performing partial self-consistency schemes [30,18,31–33,19], introducing a restriction of orbital space (e.g. by excluding most of the virtual orbitals [34]); approximating the self-energy matrix by excluding off-diagonal elements (quasiparticle approximation) [24,35]; restricting frequencies considered (by introducing plasmon-pole [36–39,19,40,41] or static approximations [42,43]).

The difficulty in executing the Green's function framework fully ab-initio is exemplified by the fact that while initial applications to model Hamiltonians describing systems as complicated as oxides started in the 1970s and were in the full swing delivering approximate descriptions of semiconductors in the 1980s [19], the fully self-consistent GW method without any approximations for electron gas was only executed in the 1997 [44].

This slow progress towards reaching fully ab-initio Green's function results for large realistic problems can be rationalized after considering the challenges present in realistic periodic systems such as (i) a wide Hamiltonian spectrum resulting in large frequency (time) grids when both core and virtual orbitals necessitate description, (ii) the difficulty of performing self-consistent iterations (resulting in an evaluation of bold diagrammatic series) (iii) the high computational scaling of weakly correlated methods such as GW and second order Green's function (GF2) scaling as $\mathcal{O}(n^6)^2$ and $\mathcal{O}(n^5)$, respectively, where n is the number of orbitals in the problem, (iv) lack of numerical tools (up to 1990s) such as Cholesky decomposition [45, 46], density fitting [46–49], the pseudospectral (PS) method [50–59], tensor hypercontraction (THC) [60,61], or Tensor Network method [62] for two-body Coulomb integrals enabling a significant reduction of computational scaling without resorting to uncontrolled diagrammatic series truncations or approximations.

The fully ab-initio, rigorous evaluation of the many-body Green's function methods without any ad hoc approximations and with controllable accuracy for large realistic systems is only now becoming possible. This is due to the substantial method development that happened within last 15 years ranging from adaptive grids [63–67], decompositions of Coulomb integrals that proved to be accurate enough for Green's function methods while reducing the computational cost by orders of magnitude, convergence acceleration schemes [68,69], development of full self-consistency in finite temperature Green's function approaches, and GPU acceleration that was necessary to enable such simulations routinely and reliably.

Almost all practical diagrammatic self-consistent methods are implemented in the finite-temperature Matsubara framework (on the imaginary axis) necessitating the use of analytic continuation methods to obtain spectral information such as band gap and band structure. While traditional methods either smooth data (such as the Maximum Entropy method [70]), or possibly violate causality (such as the Padé method [71]), recent theoretical progress resulting in the Nevanlinna [72,73], projection-estimation-semidefinite relaxation (PES) [74] and Prony [75–77] methods enables causal, accurate, and systematically improvable continuations.

While many publicly available codes evaluating weakly correlated Green's functions such as GW are available, very few codes enable a fully ab-initio rigorous evaluating of Green's function containing the newest developments described above and providing a good control of the accuracy while being available in readily accessible user-friendly simulation packages. *Green/WeakCoupling* aims to provide an implementation of these methodologies as a permissively licensed open-source software package, together with tutorials, documentation, and examples, to enable diagrammatic materials calculations with self-consistent techniques, along with additional post-processing tools.

2. Theory

Green/WeakCoupling provides an approximate solution for realistic quantum systems of interacting electrons using Green's function based many-body perturbation theory methods. The interacting quantum system is described by a second quantized Hamiltonian of the form

$$H = \sum_{ij,\sigma} H_{ij}^{\mathbf{k}_i\mathbf{k}_j(0)} c_{\mathbf{k}_i\sigma}^\dagger c_{\mathbf{k}_j\sigma} + \frac{1}{2N_k} \sum_{ijkl,\sigma\sigma'} U_{ijkl}^{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_k\mathbf{k}_l} c_{\mathbf{k}_i\sigma}^\dagger c_{\mathbf{k}_k\sigma'}^\dagger c_{\mathbf{k}_l\sigma'} c_{\mathbf{k}_j\sigma}. \quad (1)$$

² GW scaling of $\mathcal{O}(n^6)$ is present in the finite temperature Matsubara formalism when all the elements (diagonal and off-diagonal) of self-energy are evaluated and no decomposition of two-body integrals is employed. With the density fitted or Cholesky decomposed integrals, this scaling drops to $\mathcal{O}(n^4)$. A further reduction in scaling can be achieved when only diagonal elements of self-energy are evaluated.

Here, the operators $c^\dagger$ (c) create (destroy) electrons with spin σ , momentum $\mathbf{k}_i(\mathbf{k}_j, \mathbf{k}_k, \mathbf{k}_l)$ and the atomic orbital index $i(j, k, l)$ [78]. For molecular systems momentum index is omitted. Given a set of single-particle basis states $\psi_i^{\mathbf{k}_i}(\mathbf{r})$ with an overlap integral $S_{ij}^{\mathbf{k}_i, \mathbf{k}_j} = \int d\mathbf{r} \psi_i^{\mathbf{k}_i*}(\mathbf{r}) \psi_j^{\mathbf{k}_j}(\mathbf{r})$, the one-body Hamiltonian $H_{ij}^{\mathbf{k}_i, \mathbf{k}_j(0)}$, in the simplest case, is defined as

$$H_{ij}^{\mathbf{k}_i, \mathbf{k}_j(0)} = \int d\mathbf{r} \psi_i^{\mathbf{k}_i*}(\mathbf{r}) \left(-\frac{\hbar^2}{2m} \nabla_{\mathbf{r}}^2 - V_{ext}(\mathbf{r}) \right) \psi_j^{\mathbf{k}_j}(\mathbf{r}), \quad (2)$$

and describes the motion of electrons in the external potential $V_{ext}(\mathbf{r})$. The electron-electron repulsion³ is

$$U_{ijkl}^{\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_k, \mathbf{k}_l} = \int d\mathbf{r}_1 d\mathbf{r}_2 \psi_i^{\mathbf{k}_i*}(\mathbf{r}_1) \psi_j^{\mathbf{k}_j}(\mathbf{r}_1) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_k^{\mathbf{k}_k*}(\mathbf{r}_2) \psi_l^{\mathbf{k}_l}(\mathbf{r}_2). \quad (3)$$

This rank-four Coulomb tensor is typically decomposed with the help of the density fitting [47,46,48,49] or Cholesky decomposition [46,45] techniques into a product of two low-rank tensors as

$$U_{ijkl}^{\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_k, \mathbf{k}_l} = \sum_Q \tilde{V}_{\mathbf{k}_i, \mathbf{k}_j, Q} \tilde{V}_{\mathbf{k}_k, \mathbf{k}_l, Q}, \quad (4)$$

where Q is an auxiliary index. The current version of `Green/WeakCoupling` uses PySCF [79] package to prepare one- and two-body integrals and we assume that single-particle basis functions $\psi_i^{\mathbf{k}_i}(\mathbf{r})$ are either periodic Gaussian-type orbitals (for solids) or Gaussian-type orbitals (for molecules).

`Green/WeakCoupling` provides an approximate solution of system described by the Hamiltonian from Eq. (1) within the Green's function language, where the main objects are imaginary-time or Matsubara-frequency-dependent electron Green's functions and electron self-energies expressed in terms of Feynman diagrams [80], that in Matsubara frequency space are connected via the Dyson equation

$$G_{ij}^{-1}(\omega_n) = [(i\omega_n + \mu)S_{ij} - H_{ij}^0] - \Sigma_{ij}(\omega_n), \quad (5)$$

with non-interacting Green's function $G_{ij}^0(\omega_n) = [(i\omega_n + \mu)\mathbf{S} - \mathbf{H}^0]_{ij}^{-1}$.

It is convenient to split the self-energy into static and time-dependent parts:

$$\Sigma_{ij}(\tau) = \Sigma_{ij}^\infty \delta_\tau + \tilde{\Sigma}_{ij}(\tau), \quad (6)$$

where Σ_{ij}^∞ corresponds to the static infinite-frequency limit of the self-energy, and the dynamical part $\tilde{\Sigma}_{ij}(\tau)$ encapsulates the dependence on imaginary time τ , $0 \leq \tau \leq \beta$, where $\beta = 1/k_B T$ is the inverse of the physical temperature T .

The solution of the lowest-order approximation of diagrammatic perturbation theory, Hartree-Fock, leads only to a static self-energy

$$\Sigma_{\sigma, ij}^{\infty, \mathbf{k}} = \frac{1}{N_k} \sum_{\mathbf{k}', \sigma'} \tilde{V}_{i\mathbf{k}, j\mathbf{k}}(Q) G_{\sigma', ab}^{\mathbf{k}', (0-)} \tilde{V}_{b\mathbf{k}', a\mathbf{k}'}(Q) - \frac{1}{N_k} \sum_{\mathbf{k}', Q} \tilde{V}_{i\mathbf{k}, a\mathbf{k}'}(Q) G_{\sigma, ab}^{\mathbf{k}', (0-)} \tilde{V}_{b\mathbf{k}', j\mathbf{k}}(Q), \quad (7)$$

where $G_{\sigma, ij}^{\mathbf{k}}(\tau)$ is the single-particle imaginary time Green's function.

Higher order approximations yield additional dynamical corrections. `Green/WeakCoupling` provides an implementation of two approximations beyond Hartree-Fock, namely the second-order self-consistent perturbation theory (GF2) and self-consistent GW method (scGW).

2.1. GF2 approximation

GF2, also known as Second-order Born approximation [81], is a self-consistent conserving diagrammatic approximation that contains all the self-energy diagrams up to a second order in the interaction [82–84]. Generally, it is considered accurate whenever gaps are large and interactions are weak but it is known to fail for metals. Unlike GW [24] (cf. Sec. 2.2), GF2 is expanded in terms of bare Coulomb interactions, and it contains a second-order exchange term but does not include higher-order screening contributions.

The second-order self-energy in the imaginary time and momentum space with decomposed Coulomb interaction is

$$\begin{aligned} \Sigma_{\sigma, ij}^{\mathbf{k}, (2)}(\tau) = & -\frac{1}{N_k^3} \sum_{\substack{\mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3 \\ klmnpq \\ \sigma', QQ'}} (\tilde{V}_{qj}^{\mathbf{k}_1 \mathbf{k}}(Q') \tilde{V}_{ln}^{\mathbf{k}_2 \mathbf{k}_3}(Q') - \tilde{V}_{lj}^{\mathbf{k}_2 \mathbf{k}}(Q') \tilde{V}_{qn}^{\mathbf{k}_1 \mathbf{k}_3}(Q')) \delta_{\sigma\sigma'} \\ & \times \tilde{V}_{ip}^{\mathbf{k} \mathbf{k}_1}(Q) \tilde{V}_{mk}^{\mathbf{k}_3 \mathbf{k}_2}(Q) G_{\sigma, pq}^{\mathbf{k}_1}(\tau) G_{\sigma', kl}^{\mathbf{k}_2}(\tau) G_{\sigma', nm}^{\mathbf{k}_3}(-\tau) \delta_{\mathbf{k}+\mathbf{k}_3, \mathbf{k}_1+\mathbf{k}_2}, \end{aligned} \quad (8)$$

with imaginary time Green's function $G_{\sigma, ij}^{\mathbf{k}}(\tau)$.

A detailed discussion on the GF2 approximation implemented in `Green/WeakCoupling` is presented in Ref. [83,84].

2.2. GW approximation

The GW method sums an infinite series of so-called random phase approximation (RPA) or “bubble” diagrams, thereby including screening effects but neglecting the second-order exchange diagrams [24]. Note that there are several variants of the GW approximation that correspond to different equations and additional approximations, including non-selfconsistent [19], partially self-consistent [44], quasiparticle approximated and quasiparticle self-consistent variants [85]. `Green/WeakCoupling` implements fully self-consistent GW approximation [86,87].

³ `Green/WeakCoupling` uses chemists notation (see Ref. [78]) for the electron-electron repulsion, as it's commonly used in many ab initio packages.

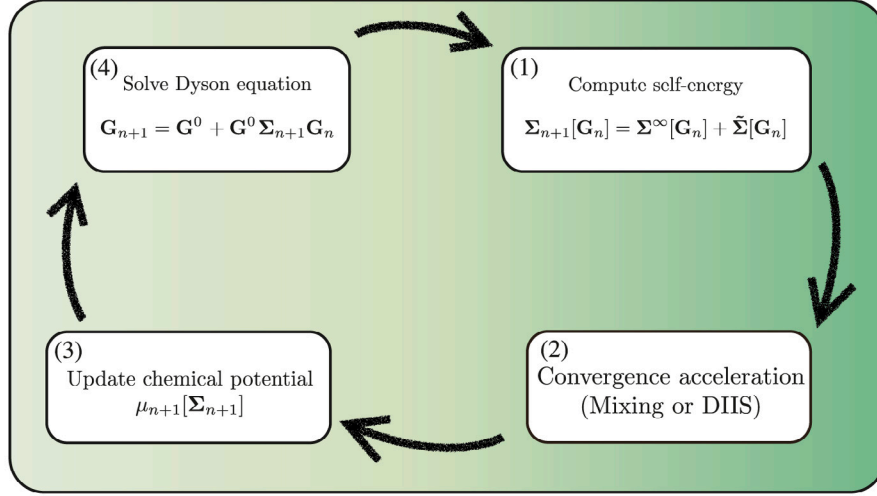


Fig. 1. Schematic illustration of the self-consistency loop implemented in Green/WeakCoupling.

In scGW, the contribution to the dynamical part of the self-energy is

$$\tilde{\Sigma}_{i\sigma,j\sigma}^{(GW),\mathbf{k}}(\tau) = -\frac{1}{N_k} \sum_{\mathbf{q}} \sum_{ab} G_{\sigma,ab}^{\mathbf{k}-\mathbf{q}}(\tau) \tilde{W}_{iabj}^{\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{k}}(\tau), \quad (9)$$

with dynamical screened interaction expressed in the frequency space as

$$\tilde{W}_{ijkl}^{\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{k}}(i\Omega_n) = \sum_{QQ'} \tilde{V}_{ij}^{\mathbf{k},\mathbf{k}-\mathbf{q}}(Q) \tilde{P}_{QQ'}^{\mathbf{q}}(i\Omega_n) \tilde{V}_{kl}^{\mathbf{k}-\mathbf{q},\mathbf{k}}(Q') \quad (10)$$

where $\Omega_n = 2n\pi/\beta$ ($n \in \mathbb{Z}$) are bosonic Matsubara frequencies. $\tilde{P}_{QQ'}^{\mathbf{q}}(i\Omega_n)$ is a renormalized auxiliary function

$$\tilde{\mathbf{P}}^{\mathbf{q}}(i\Omega_n) = \sum_{m=1}^{\infty} [\tilde{\mathbf{P}}_0^{\mathbf{q}}(i\Omega_n)]^m = [\mathbf{I} - \tilde{\mathbf{P}}_0^{\mathbf{q}}(i\Omega_n)]^{-1} \tilde{\mathbf{P}}_0^{\mathbf{q}}(i\Omega_n), \quad (11)$$

that is obtained from a bare auxiliary function

$$\tilde{P}_{0,QQ'}^{\mathbf{q}}(i\Omega_n) = \int_0^{\beta} d\tau \tilde{P}_{0,QQ'}^{\mathbf{q}}(\tau) e^{i\Omega_n \tau}, \quad (12)$$

$$\tilde{P}_{0,QQ'}^{\mathbf{q}}(\tau) = -\frac{1}{N_k} \sum_{\mathbf{k}\sigma} \tilde{V}_{da}^{\mathbf{k},\mathbf{k}+\mathbf{q}}(Q) G_{\sigma,cd}^{\mathbf{k}}(-\tau) G_{\sigma,ab}^{\mathbf{k}+\mathbf{q}}(\tau) \tilde{V}_{bc}^{\mathbf{k}+\mathbf{q},\mathbf{k}}(Q') \quad (13)$$

For detailed derivation of the GW formalism implemented within Green/WeakCoupling with a non-relativistic Hamiltonian refer to Ref. [86] A relativistic GW (X2C1e-scGW) within the exact two-component formalism (X2C1e) [88] is presented in Ref. [89].

2.3. Self-consistency loop

Green/WeakCoupling provides an implementation of the fully self-consistent many-body perturbation theory. Self-consistent iterations start from an initial guess for the self-energy Σ , usually either obtained from a ground-state mean-field solution, from previous calculations or set to zero for the non-interacting limit, and proceed as following. (0) A trial Green's function is evaluated (Eq. (5)). (1) The static part (Eq. (7)) and the dynamic part (Eq. (8) or Eq. (9)) of the self-energy are evaluated. (2) To improve convergence, a convergence acceleration technique is applied. (3) The chemical potential is adjusted to obtain a fixed number of particles. (4) A new approximation for the Green's function is calculated via the Dyson equation (Eq. (5)) and the simulation proceeds to step (1) until the desired accuracy of convergence has been reached. Fig. 1 shows a schematic representation of the aforementioned procedure.

2.4. Postprocessing

2.4.1. k-path interpolation

Green/WeakCoupling evaluates Green's functions and self-energies on a relatively small Monkhorst-Pack $\mathbf{k}$ -grid [90]. However, a visually appealing presentation of quantities such as the band-structure requires a dense $\mathbf{k}$ -mesh.

The frequency-momentum representation of the Green's function obtained by Green/WeakCoupling is

$$G_{ij}^{\mathbf{k}}(\omega_n) = [(i\omega_n + \mu)\mathbf{S}^{\mathbf{k}} - \mathbf{H}^{\mathbf{k},(0)} - \Sigma^{\infty,\mathbf{k}} - \tilde{\Sigma}^{\mathbf{k}}(\omega_n)]_{ij}^{-1}, \quad (14)$$

with Matsubara frequency $\omega_n = (2n + 1)\pi/\beta$, chemical potential μ , and overlap matrix $S_{ij}^{\mathbf{k}}$.

To obtain the Green's function on a denser grid $\hat{\mathbf{k}}$, we perform a simple $\mathbf{k}$ -space interpolation similar to a Wannier interpolation [91], with the exception that we do not perform the transformation to a basis of maximally localized Wannier functions.

In this approach, we first transform momentum dependent quantities $\hat{X}(\mathbf{k})$ into real space via Fourier transform,

$$X(\mathbf{R}) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} \hat{X}(\mathbf{k}) e^{-i\mathbf{k}\mathbf{R}}. \quad (15)$$

Then, by applying the inverse transform, we project them back onto a finer $\hat{\mathbf{k}}$ -grid

$$\hat{X}(\hat{\mathbf{k}}) = \sum_{\mathbf{R}} X(\mathbf{R}) e^{i\hat{\mathbf{k}}\mathbf{R}}. \quad (16)$$

If the real-space quantity $X(\mathbf{R})$ is well localized, which is usually the case when single-particle basis functions $\psi_i^{\mathbf{k}_i}(\mathbf{r})$ are Gaussian-type orbitals, the resulting quantity will be approximated well. We found that the best results are obtained when only $\Sigma^{\infty, \mathbf{k}}$ and $\hat{\Sigma}^{\mathbf{k}}$ are interpolated, and both the non-interacting Hamiltonian H^0 and the overlap matrix S are directly evaluated on a finer $\hat{\mathbf{k}}$ -grid. Then the Green's function is evaluated using the Eq. (14) on a finer $\hat{\mathbf{k}}$ -grid.

2.4.2. Analytical continuation

Standard finite-temperature perturbation theories are formulated on the imaginary axis allowing the direct access to static quantities such as the density matrix, free energy, or entropy. In contrast, quantities defined on real-frequency grids such as the spectral function require analytical continuation to the real axis of imaginary-time data [70,92] by solving the inverse problem of

$$G_{ij}^{\sigma}(\tau) = - \int d\omega \frac{A_{ij}^{\sigma}(\omega) e^{-\tau\omega}}{1 + e^{-\beta\omega}}. \quad (17)$$

The solution of the analytical continuation problem is ill conditioned and numerically unstable in the presence of noise [70]. *Green/WeakCoupling* results are free of stochastic noise and methods suitable for noise-free data can be applied. *Green/WeakCoupling* provides an implementation of analytical continuation using the Nevanlinna analytical continuation method as described in Ref. [72]. Several implementations of this method exist [93,94].

3. Installation

3.1. Dependencies

Green requires the following dependencies to be installed and available:

- C++17 compatible compiler.
- Eigen3 >= 3.4.0. Eigen is a C++ template library for linear algebra: matrices, vectors, numerical solvers, and related algorithms [95].
- MPI >= 3.0. Message Passing Interface (MPI) is a standardized and portable message-passing standard designed to function on parallel computing architectures [96].
- HDF5 >= 1.10.0. HDF5 is a high-performance data management and storage suite [97].
- BLAS. The BLAS (Basic Linear Algebra Subprograms) are routines that provide standard building blocks for performing basic vector and matrix operations [98].
- CMake >= 3.18. CMake is a tool to manage building of source code [99].
- GNU Make [100] or Ninja build system [101].

The following packages are optional and needed if certain modules of the *Green* software package are included:

- CUDAToolkit >= 11.1 (needed for the GPU kernel). CUDA is a parallel computing platform and programming model for general computing on graphical processing units (GPUs) [102].
- GMP (needed for the analytic continuation module). The GNU Multiple Precision Arithmetic Library (GMP) is a library for arbitrary-precision arithmetic, operating on signed integers, rational numbers, and floating-point numbers [103].

These packages are external to *Green*. Installation instructions are provided with these packages; many modern HPC environments provide some or all of these packages preinstalled for their users.

In addition, the following python dependencies are required for auxiliary scripts.

- green-mbtools. The python tool suite for *Green*'s-function-based many-body calculations using *Green* Software Package.
- PySCF. The Python-based Simulations of Chemistry Framework (PySCF) is an open-source collection of electronic structure modules powered by Python [79].
- numba. Numba translates Python functions to optimized machine code at runtime using the industry-standard LLVM compiler library [104].
- spglib. SPGLib is a software library for crystal symmetry search [105].
- ase. ASE is an Atomic Simulation Environment written in the Python programming language with the aim of setting up, steering, and analyzing atomistic simulations [106].

Python packages can typically be provisioned with the `pip install` or `pip3 install` command. `pip install pyscf numba spglib ase green-mbtools` will install the required python modules on most platforms.

3.2. Weak coupling

The following instructions will download, build and install the CPU-only version of Green/WeakCoupling solver (replace /absolute/path/to/install/directory with the desired installation directory path; see below for the CPU/GPU version):

```
$ git clone https://github.com/Green-Phys/green-mbpt
$ cmake -S green-mbpt -B green-mbpt-build -DCMAKE_BUILD_TYPE=Release \
    -DCMAKE_INSTALL_PREFIX=/absolute/path/to/install/directory
$ cmake --build green-mbpt-build -j 4
$ cmake --build green-mbpt-build --target test install
$ rm -rf green-mbpt-build
```

This sequence of commands will create the installation directory and place the executable mbpt.exe under the bin directory of the installation path.

The following instructions will download, build and install the GPU version of the Green/WeakCoupling solver (replace /absolute/path/to/install/directory with the desired installation directory path):

```
$ git clone https://github.com/Green-Phys/green-mbpt
$ cmake -S green-mbpt -B green-mbpt-build -DCMAKE_BUILD_TYPE=Release \
    -DCMAKE_INSTALL_PREFIX=/absolute/path/to/install/directory \
    -DCUSTOM_KERNEL=GPU_KERNEL \
    -DGREEN_KERNEL_URL="https://github.com/Green-Phys/green-gpu" \
    -DGREEN_CUSTOM_KERNEL_LIB="GREEN::GPU" \
    -DGREEN_CUSTOM_KERNEL_ENUM=GPU \
    -DGREEN_CUSTOM_KERNEL_HEADER("<green/gpu/gpu_factory.h>")
$ cmake --build green-mbpt-build -j 4
$ cmake --build green-mbpt-build --target test install
$ rm -rf green-mbpt-build
```

The GPU version requires the CUDA toolkit [102] to be available.

3.3. Analytical continuation

In addition to the Green/WeakCoupling dependencies, the analytical continuation code requires the GNU multiprecision arithmetics library [103]. The following instructions will download and build the analytical continuation package (replace /absolute/path/to/install/directory with the desired installation directory):

```
$ git clone https://github.com/Green-Phys/green-ac
$ cmake -S green-ac -B green-ac-build \
    -DCMAKE_INSTALL_PREFIX=/absolute/path/to/install/directory \
    -DCMAKE_BUILD_TYPE=Release
$ cmake --build green-ac-build -j 4
$ cmake --build green-ac-build -j 4 --target test install
$ rm -rf green-ac-build
```

4. Usage

4.1. Input preparation

The first step of any simulation with Green/WeakCoupling is the preparation of the input data for the diagrammatic calculation. This entails the calculation of the one-body integrals, the calculation of the (decomposed or density fitted) Coulomb integrals, and the determination of a starting density matrix. These components are generated from unit-cell geometry information, atom positions, the k-point discretization grid, as well as information about the basis.

We provide a convenient interface to the PySCF package [79] to prepare this input by calling the python script python/init_data_df.py, located in the installation path (for a simple example see section 6). The following parameters are mandatory:

- --a – path to a file containing the translation vectors of the primitive unit cell, in angstrom.
- --atom – path to a file containing the atom species and position of atoms in the unit cell in the standard xyz format, in angstrom.
- --nk – number of k-points in each direction. Assumed to be the same for all directions.
- --basis – Gaussian basis set description.

The Green/WeakCoupling interface to pySCF employs Gaussian-type orbitals as evaluated by PySCF [79] as single-particle states. Two options for choosing a basis set are provided: a) using a built-in molecular [107] or periodic [108] basis; b) providing a file that contains desired basis set in the NWChem [109] format. Users can either select a single basis set for all atoms in the calculation (--basis <basis set>) or provide an individual basis for each atom species separately:

```
--basis <first atom> <first basis set> <second atom> <second basis>...
```

Basis sets with pseudopotential require the user to provide a pseudopotential for core electrons using the option --pseudo. Users can either choose a built-in PySCF pseudopotential [110] or provide a file containing pseudopotentials in the NWChem format.

Users can use an external MolSSI BSE tool [111] for basis set conversions from other formats.

By default the script `init_data_df.py` will generate a file `input.h5` containing all necessary parameters and an initial mean-field solution of the system, as well as the one-body integrals. In addition, the script will generate a directory named `df_hf_int` which contains the decomposed Coulomb integrals of the system with integrable divergence at $(\mathbf{q} + \mathbf{G}) = 0$ explicitly removed. If parameter `--finite_size_kind` set to `ewald` script will generate an additional directory named `df_int` which contains the decomposed Coulomb integrals with integrable divergence at $(\mathbf{q} + \mathbf{G}) = 0$ replaced with an estimate using Madelung constant (see Sec. 4.2.1 for more details on finite-size corrections).

After the simulation is performed, some of the results, such as the single particle spectral function, are displayed along a high-symmetry path. To obtain a high-symmetry path, high-symmetry points on the path and the total number of points along the path are provided using the two parameters `--high_symmetry_path` and `--high_symmetry_path_points`. The option `--print_high_symmetry_points` lists all available high-symmetry points for a system. The interpolation along the desired high-symmetry paths will perform a basic $\mathbf{k}$ -path interpolation (see Sec. 2.4.1) for those points not on the momentum mesh of the calculation.

All Green programs list their available options with the `--help` option.

4.2. Solution of the many-body perturbation theory equations

The many-body perturbation theory equations are solved by the program `mbpt.exe` located in the `bin` directory of the installation path. It is advantageous to perform this simulation on a machine with multiple cores or nodes (using the CPU MPI implementation) or on GPUs (using the GPU implementation), rather than on a local workstation or laptop. Minimal parameters needed to run weak-coupling simulations are as follows:

- `--BETA` – The inverse temperature, in inverse energy units
- `--scf_type` – The type of the diagrammatic approximation, either HF (for Hartree Fock), GF2 (for fully self-consistent second-order perturbation theory), or GW (for fully self-consistent GW)
- `--grid_file` – the path to a file containing the non-uniform grid information. The program will automatically check three possible locations in the following order:
 - current directory or absolute path
 - `<installation directory>/share`
 - the build directory of mbpt code

As a default, we provide intermediate representation (IR [66], `ir` subdirectory) and Chebyshev ([65], `cheb` subdirectory) grids for nonuniform imaginary time representation.

After successful completion, results will be written to a file located at `--results_file` (by default set to `sim.h5`) Additional parameters and their default values are listed by calling the program with the `--help` option.

Provided the option `--jobs WINTER` is set and the input data contains information about a desired high-symmetry path, the diagonal part of the Green's function is evaluated on the path, and results are stored in the `G_tau_hs` group in the file specified as `--high_symmetry_output_file` (by default set to `output_hs.h5`).

4.2.1. Finite-size corrections

In periodic calculations the infinite crystal is approximated by a finite crystal with periodic boundary conditions defined on a discrete set of momentum points. This finite-size approximation introduces finite-size errors. *Green/WeakCoupling* provides the following options to control finite-size errors:

In Hartree-Fock calculations we compute explicit Ewald corrections using the Madelung constant following Baldereschi's scheme [112].

In GW calculations we provide two options:

- `EWALD_INT` enables explicit Ewald correction using integrals with divergent part of Coulomb kernel replaced with supercell Madelung constant [113,114];
- `EXTRAPOLATE` computes an extrapolation of the dielectric matrix to obtain the $\mathbf{G} = 0$ contribution. This requires the calculation of a smooth interpolant for the Coulomb integral [113,114,86].

In the GF2 calculations, the divergent part of the Coulomb kernel is replaced with the supercell Madelung constant via the Ewald correction [113, 114]. The finite-size corrections will be applied if the file `df_ewald.h5` is present in the integrals directory.

The finite-size corrections for the second-order and GW approximations require the generation of additional input data. In the initialization script, this is controlled by the `--finite_size_kind` parameter which takes the following values

- `ewald` – will generate the `df_int` directory which contain the additional set of integrals with explicit Ewald correction;
- `gf2` – will generate the file `df_ewald.h5` that contains Ewald correction for integrals in second-order diagrams.
- `gw` – will generate the file `AqQ.h5` that contains a smooth interpolant for Coulomb integrals for the GW finite-size correction.

By default the `ewald` correction is applied.

4.2.2. Convergence to a desired solution and convergence acceleration

Self-consistent diagrammatic theories usually display multiple stationary points in their Φ - and Ψ -functional and very rarely have only one global minimum. In certain cases, the existence of these multiple stationary points may be employed to our advantage since giving a starting solution that lies in the vicinity of the desired stationary point (local minimum) will predispose the algorithm to converge to this solution. This is usually done by searching for a desired solution in the mean-field (Hartree-Fock) method that produces multiple solutions, known as broken-symmetry solutions [115]. Subsequently, the weakly correlated perturbative fully self-consistent diagrammatic method often preserves the qualitative structure

of the mean-field solution, thus, providing access to a broken-symmetry solution of the Dyson equation. We have successfully used these correlated solutions for accurate evaluation of magnetic properties of molecules [13] and solids [116] and even critical temperatures [117].

To enable a smooth convergence to a desired solution and minimize the number of iterations required, its convergence path may be altered either by a constant iteration mixing, or by convergence acceleration methods such as variants of the direct inversion in the iterative subspace (DIIS) [118,119,68].

In iteration mixing, the self-energy is modified as:

$$\Sigma_n = \alpha \Sigma [G_{n-1}] + (1 - \alpha) \Sigma_{n-1}, \quad (18)$$

where G_{n-1} and Σ_{n-1} are Green's function and self-energy from the previous iteration and $\alpha \in (0, 2)$ is an iteration mixing parameter (with $\alpha = 1$ meaning no mixing with previous iteration).

In DIIS, the self-energy at the given iteration is computed as a linear combination of the self-energies obtained in the k previous iterations:

$$\Sigma_n = \sum_{i=0}^k c_k \Sigma_{n-1-k+i}. \quad (19)$$

Convergence acceleration methods often offer faster convergence and better stability.

To choose a convergence acceleration method, the user has to specify the parameter `--mixing_type` which takes one of the four values `NO_MIXING`, `SIGMA_MIXING`, `DIIS`, or `CDIIS`. Where `CDIIS` corresponds to commutator version of DIIS method [119]. If static iteration mixing is specified, the mixing weight α has to be provided by specifying `--mixing_weight`. If DIIS or CDIIS are chosen, the `Green/WeakCoupling` code uses static iteration mixing for the first few iterations, builds an extrapolation subspace, and continues with DIIS/CDIIS starting at the iteration specified by `--diis_start`. The maximum number of subspace vectors used is specified by `--diis_size`. Our implementation of DIIS and CDIIS is detailed in Ref. [68].

5. Post processing and analysis

The `Green/WeakCoupling` code provides post-processing tools such as analytical continuation to obtain spectral functions and thermodynamic utilities.

Analytical continuation using the Nevanlinna analytic continuation method [72] is performed by executing the program `ac.exe` located at the installation path in the `bin` subdirectory. The following parameters are needed:

- `--grid_file` Sparse imaginary time/frequency grid file name
- `--BETA` Inverse temperature
- `--input_file` Name of the input file
- `--output_file` Name of the output file
- `--group` Name of the HDF5 group in the input file that contains imaginary time data. This group has to contain mesh information and data sets.
- `--kind` Type of analytical continuation, currently only `NEVANLINNA` is available.

The command `ac.exe --help` will provide additional information.

6. Example

We give step-by-step instructions on how to run `Green/WeakCoupling` for periodic silicon on a $6 \times 6 \times 6$ lattice. Results for a similar system are shown in Ref. [86]. Both `mbpt.exe` and `ac.exe` will be used.

Create a new simulation directory, and in it create the file `a.dat` containing the following unit cell information (each line specifies one of the translation vectors in Angstroms):

```
0.0000, 2.7155, 2.7155
2.7155, 0.0000, 2.7155
2.7155, 2.7155, 0.0000
```

Then create a file `atom.dat` containing atom positions in cell Angstroms unit cell:

```
Si 0.00000 0.00000 0.00000
Si 1.35775 1.35775 1.35775
```

In a first step, obtain input parameters and an initial mean-field solution by running `pySCF` via the `init_data_df.py` script:

```
python <source root>/green-mbpt/python/init_data_df.py \
--a a.dat --atom atom.dat --nk 6 \
--basis gth-dzvp-molopt-sr --pseudo gth-pbe --xc PBE \
--high_symmetry_path WGXWLG --high_symmetry_path_points 300 \
--job init sym_path
```

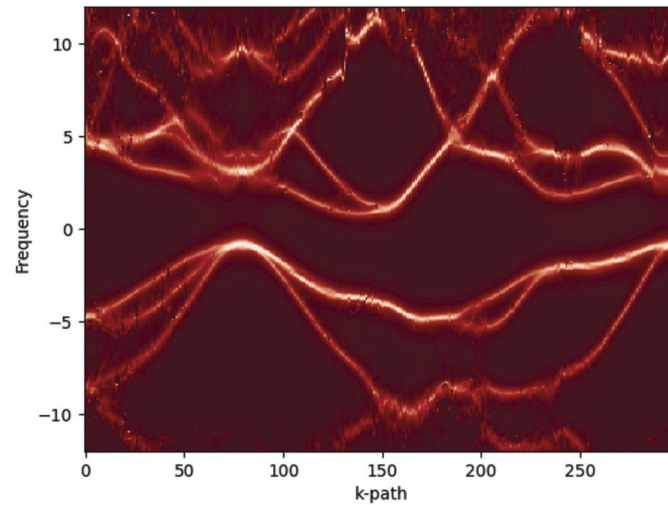



Fig. 2. Band structure for silicon obtained from GW calculation on $6 \times 6 \times 6$ lattice and extrapolated along high-symmetry path.

This will employ the `gth-dzvp-molopt-sr` basis [120] with the `gth-pbe` pseudopotential, run a DFT calculation for the system with a PBE exchange correlation potential⁴ and generate Green/WeakCoupling input. The high-symmetry path, which will be used in the analysis step, is set to `WGXWLG`.

In a second step, we run the fully self-consistent GW approximation using the `mbpt.exe` program. It is advantageous to run this step on an MPI cluster or on a GPU:

```
mpirun -n <number of cores> <install dir>/bin/mbpt.exe --scf_type=GW --BETA 100 \
--grid_file ir/1e4.h5 --itermax 20 --results_file Si.h5 \
--mixing_type CDIIS --diis_size 5 --diis_start 2 \
--high_symmetry_output_file Si_hs.h5 --jobs SC,WINTER
```

This sets the inverse temperature to $\beta = 100 \text{ Ha}^{-1}$, employs an Intermediate Representation grid [66,121] with grid parameter $\Lambda = 10^4$, and runs for 20 iterations using DIIS convergence acceleration.

The simulation results are stored into the file `Si.h5`, and the band-path interpolation along the high-symmetry path in `Si_hs.h5`.

In a third step, we use the post-processing analytic continuation to obtain the spectral function:

```
<install dir>/bin/ac.exe --BETA 100 --grid_file ir/1e4.h5 \
--input_file Si_hs.h5 --output_file ac.h5 --group G_tau_hs \
--e_min -10.0 --e_max 10.0 --n_omega 10000 --eta 0.01
--kind Nevanlinna
```

This will run analytical continuation for all momentum points along the high-symmetry path. The output will be stored in the group `G_tau_hs` of the output HDF5 file `ac.h5`.

Finally, a plot for the band structure can be obtained with the `plot_bands.py` script:

```
python <source root>/green-mbpt/python/plot_bands.py \
--input_file ac.h5 --output_dir bands
```

This will read the analytically continued data and plot it to an `<output_dir>/bands.png` file. In addition, it will create a plain-text data file containing the spectral function for every k-point along the chosen path inside the `<output_dir>` directory. The resulting band structure plot is shown in Fig. 2.

Other examples, such as calculation of ionization potentials in molecular systems [122] or using spin-orbit coupling [123], please check the Green/WeakCoupling web-site.

7. Conclusion

We have presented an implementation of self-consistent finite-temperature many-body perturbation theory for molecules and solids, along with analytic continuation tools. The software provided as part of the Green project provides a way to perform diagrammatic simulations of solids and molecules without adjustable parameters. We have provided scripts and examples for software installation, input preparation, solution of the many-body problem, and analytic continuation analysis. Our implementation is licensed with the permissive MIT license that allows users to use, copy, modify, merge, publish, distribute, sublicense, and sell copies of software.

⁴ If the `--xc` parameter is omitted, the script will run a Hartree-Fock calculation instead.

CRediT authorship contribution statement

Sergei Iskakov: Writing – review & editing, Writing – original draft, Visualization, Supervision, Software, Project administration. **Chia-Nan Yeh:** Writing – review & editing, Software. **Pavel Pokhilko:** Writing – review & editing, Software. **Yang Yu:** Writing – review & editing, Validation. **Lei Zhang:** Software. **Gaurav Harsha:** Software. **Vibin Abraham:** Software. **Ming Wen:** Validation. **Munkhorgil Wang:** Validation. **Jacob Adamski:** Validation. **Tianran Chen:** Validation. **Emanuel Gull:** Writing – review & editing, Writing – original draft, Supervision, Project administration. **Dominika Zgid:** Writing – review & editing, Writing – original draft, Validation, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This material is based upon work supported by the National Science Foundation under Grant No. OAC-2310582.

References

- [1] P. Hohenberg, W. Kohn, Inhomogeneous electron gas, *Phys. Rev.* 136 (1964) B864–B871, <https://doi.org/10.1103/PhysRev.136.B864>.
- [2] W. Kohn, L.J. Sham, Self-consistent equations including exchange and correlation effects, *Phys. Rev.* 140 (1965) A1133–A1138, <https://doi.org/10.1103/PhysRev.140.A1133>.
- [3] A. Abrikosov, L. Gorkov, I. Dzyaloshinski, R. Silverman, *Methods of Quantum Field Theory in Statistical Physics*, Dover Books on Physics, Dover Publications, 2012, <https://books.google.com/books?id=JYTCAgAAQBAJ>.
- [4] A.L. Fetter, J.D. Walecka, *Quantum Theory of Many-Particle Systems*, Courier Corporation, 2003.
- [5] G. Mahan, *Many-Particle Physics*, Physics of Solids and Liquids, Springer, 2000, <https://books.google.com/books?id=xzSgZ4-yyMEC>.
- [6] R.M. Martin, L. Reining, D.M. Ceperley, *Interacting Electrons: Theory and Computational Approaches*, 1st edition, Cambridge University Press, Cambridge, 2016.
- [7] W.H. Dickhoff, D. Van Neck, *Many-Body Theory Exposed!: Propagator Description of Quantum Mechanics in Many-Body Systems*, World Scientific, 2008.
- [8] B. Holm, F. Aryasetiawan, Total energy from the galitskii-migdal formula using realistic spectral functions, *Phys. Rev. B* 62 (2000) 4858–4865, <https://doi.org/10.1103/PhysRevB.62.4858>.
- [9] J.A. Sobota, Y. He, Z.-X. Shen, Angle-resolved photoemission studies of quantum materials, *Rev. Mod. Phys.* 93 (2021) 025006, <https://doi.org/10.1103/RevModPhys.93.025006>.
- [10] E.E. Salpeter, H.A. Bethe, A relativistic equation for bound-state problems, *Phys. Rev.* 84 (1951) 1232–1242, <https://doi.org/10.1103/PhysRev.84.1232>.
- [11] N.E. Bickers, S.R. White, Conserving approximations for strongly fluctuating electron systems. ii. Numerical results and parquet extension, *Phys. Rev. B* 43 (1991) 8044–8064, <https://doi.org/10.1103/PhysRevB.43.8044>.
- [12] N.E. Bickers, *Self-Consistent Many-Body Theory for Condensed Matter Systems*, Springer New York, New York, NY, 2004, pp. 237–296, Ch. 6.
- [13] P. Pokhilko, D. Zgid, Interpretation of multiple solutions in fully iterative GF2 and GW schemes using local analysis of two-particle density matrices, *J. Chem. Phys.* 155 (2) (2021) 024101, <https://doi.org/10.1063/5.0055191>.
- [14] J.M. Luttinger, J.C. Ward, Ground-state energy of a many-fermion system. ii, *Phys. Rev.* 118 (1960) 1417–1427, <https://doi.org/10.1103/PhysRev.118.1417>.
- [15] C.-O. Almbladh, U.V. Barth, R.V. Leeuwen, Variational total energies from ϕ - and ψ -derivable theories, *Int. J. Mod. Phys. B* 13 (05n06) (1999) 535–541, <https://doi.org/10.1142/S0217979299000436>.
- [16] G. Baym, L.P. Kadanoff, Conservation laws and correlation functions, *Phys. Rev.* 124 (1961) 287–299, <https://doi.org/10.1103/PhysRev.124.287>.
- [17] B. Farid, On the Luttinger-Ward functional and the convergence of skeleton diagrammatic series expansion of the self-energy for Hubbard-like models, *arXiv:2108.10903*, 2021.
- [18] M. van Schilfhaarde, T. Kotani, S. Faleev, Quasiparticle self-consistent *gw* theory, *Phys. Rev. Lett.* 96 (2006) 226402, <https://doi.org/10.1103/PhysRevLett.96.226402>.
- [19] M.S. Hybertsen, S.G. Louie, Electron correlation in semiconductors and insulators: band gaps and quasiparticle energies, *Phys. Rev. B* 34 (1986) 5390–5413, <https://doi.org/10.1103/PhysRevB.34.5390>.
- [20] A. Georges, G. Kotliar, W. Krauth, M.J. Rozenberg, Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions, *Rev. Mod. Phys.* 68 (1996) 13–125, <https://doi.org/10.1103/RevModPhys.68.13>.
- [21] D. Zgid, E. Gull, Finite temperature quantum embedding theories for correlated systems, *New J. Phys.* 19 (2) (2017) 023047, <https://doi.org/10.1088/1367-2630/aa5d34>.
- [22] G. Kotliar, S.Y. Savrasov, K. Haule, V.S. Oudovenko, O. Parcollet, C.A. Marianetti, Electronic structure calculations with dynamical mean-field theory, *Rev. Mod. Phys.* 78 (2006) 865–951, <https://doi.org/10.1103/PhysRevB.78.865>.
- [23] D. Bohm, D. Pines, A collective description of electron interactions: iii. Coulomb interactions in a degenerate electron gas, *Phys. Rev.* 92 (1953) 609–625, <https://doi.org/10.1103/PhysRev.92.609>.
- [24] L. Hedin, New method for calculating the one-particle Green's function with application to the electron-gas problem, *Phys. Rev.* 139 (1965) A796–A823, <https://doi.org/10.1103/PhysRev.139.A796>.
- [25] W.P. Reinhardt, J.D. Doll, Direct calculation of natural orbitals by many-body perturbation theory: application to helium, *J. Chem. Phys.* 50 (6) (1969) 2767–2768, <https://doi.org/10.1063/1.1671446>.
- [26] G. Csanak, H. Taylor, R. Yaris, *Green's Function Technique in Atomic and Molecular Physics*, Advances in Atomic and Molecular Physics, vol. 7, Academic Press, 1971, pp. 287–361.
- [27] F. Aryasetiawan, O. Gunnarsson, The gw method, *Rep. Prog. Phys.* 61 (3) (1998) 237, <https://doi.org/10.1088/0034-4885/61/3/002>.
- [28] L. Reining, The GW approximation: content, successes and limitations, *WIREs Comput. Mol. Sci.* 8 (3) (2018) e1344, <https://doi.org/10.1002/wcms.1344>.
- [29] D. Golze, M. Dvorak, P. Rinke, The gw compendium: a practical guide to theoretical photoemission spectroscopy, *Front. Chem.* 7 (2019), <https://doi.org/10.3389/fchem.2019.00377>.
- [30] M. van Schilfhaarde, T. Kotani, S. Faleev, Quasiparticle self-consistent *GW* theory, *Phys. Rev. Lett.* 96 (22) (2006) 226402, <https://doi.org/10.1103/PhysRevLett.96.226402>.
- [31] A. Kutepov, K. Haule, S.Y. Savrasov, G. Kotliar, Electronic structure of Pu and Am metals by self-consistent relativistic *GW* method, *Phys. Rev. B* 85 (15) (2012) 155129, <https://doi.org/10.1103/PhysRevB.85.155129>.
- [32] A.L. Kutepov, V.S. Oudovenko, G. Kotliar, Linearized self-consistent quasiparticle GW method: application to semiconductors and simple metals, *Comput. Phys. Commun.* 219 (2017) 407–414, <https://doi.org/10.1016/j.cpc.2017.06.012>.
- [33] P. García-González, R.W. Godby, Self-consistent calculation of total energies of the electron gas using many-body perturbation theory, *Phys. Rev. B* 63 (7) (2001) 075112, <https://doi.org/10.1103/PhysRevB.63.075112>.
- [34] M. Govoni, G. Galli, Large scale gw calculations, *J. Chem. Theory Comput.* 11 (6) (2015) 2680–2696, <https://doi.org/10.1021/ct500958p>, PMID: 26575564.
- [35] L. Hedin, B. Lundqvist, Explicit local exchange-correlation potentials, *J. Phys. C, Solid State Phys.* 4 (14) (1971) 2064–2083.
- [36] B.I. Lundqvist, Single-particle spectrum of the degenerate electron gas, *Phys. Kondens. Mater.* 6 (3) (1967) 193–205, <https://doi.org/10.1007/BF02422716>.
- [37] B.I. Lundqvist, Single-particle spectrum of the degenerate electron gas, *Phys. Kondens. Mater.* 6 (3) (1967) 206–217, <https://doi.org/10.1007/BF02422717>.

- [38] L. Hedin, S. Lundqvist, Effects of Electron-Electron and Electron-Phonon Interactions on the One-Electron States of Solids, *Solid State Physics*, vol. 23, Academic Press, 1970, pp. 1–181.
- [39] A.W. Overhauser, Simplified theory of electron correlations in metals, *Phys. Rev. B* 3 (1971) 1888–1898, <https://doi.org/10.1103/PhysRevB.3.1888>.
- [40] G.E. Engel, B. Farid, Generalized plasmon-pole model and plasmon band structures of crystals, *Phys. Rev. B* 47 (1993) 15931–15934, <https://doi.org/10.1103/PhysRevB.47.15931>.
- [41] P. Larson, M. Dvorak, Z. Wu, Role of the plasmon-pole model in the gw approximation, *Phys. Rev. B* 88 (2013) 125205, <https://doi.org/10.1103/PhysRevB.88.125205>.
- [42] M.S. Hybertsen, S.G. Louie, Model dielectric matrices for quasiparticle self-energy calculations, *Phys. Rev. B* 37 (1988) 2733–2736, <https://doi.org/10.1103/PhysRevB.37.2733>.
- [43] F. Bechstedt, R. Del Sole, G. Cappellini, L. Reining, An efficient method for calculating quasiparticle energies in semiconductors, *Solid State Commun.* 84 (7) (1992) 765–770, [https://doi.org/10.1016/0038-1098\(92\)90476-P](https://doi.org/10.1016/0038-1098(92)90476-P).
- [44] U. von Barth, B. Holm, Self-consistent gw_0 results for the electron gas: fixed screened potential w_0 within the random-phase approximation, *Phys. Rev. B* 54 (1996) 8411–8419, <https://doi.org/10.1103/PhysRevB.54.8411>.
- [45] H. Koch, A. Sánchez de Merás, T.B. Pedersen, Reduced scaling in electronic structure calculations using Cholesky decompositions, *J. Chem. Phys.* 118 (21) (2003) 9481–9484, <https://doi.org/10.1063/1.1578621>.
- [46] N.H.F. Beebe, J. Linderberg, Simplifications in the generation and transformation of two-electron integrals in molecular calculations, *Int. J. Quant. Chem.* 12 (4) (1977) 683–705, <https://doi.org/10.1002/qua.560120408>.
- [47] J.L. Whitten, Coulombic potential energy integrals and approximations, *J. Chem. Phys.* 58 (10) (1973) 4496–4501, <https://doi.org/10.1063/1.1679012>.
- [48] L. Maschio, Local mp2 with density fitting for periodic systems: a parallel implementation, *J. Chem. Theory Comput.* 7 (9) (2011) 2818–2830, <https://doi.org/10.1021/ct200352g>.
- [49] Š. Varga, Density fitting of Coulomb integrals in electronic structure calculations of solids, *Phys. Rev. B* 71 (2005) 073103, <https://doi.org/10.1103/PhysRevB.71.073103>.
- [50] R.A. Friesner, Solution of self-consistent field electronic structure equations by a pseudospectral method, *Chem. Phys. Lett.* 116 (1) (1985) 39–43, [https://doi.org/10.1016/0009-2614\(85\)80121-4](https://doi.org/10.1016/0009-2614(85)80121-4).
- [51] R.A. Friesner, Solution of the Hartree–Fock equations by a pseudospectral method: application to diatomic molecules, *J. Chem. Phys.* 85 (3) (1986) 1462–1468, <https://doi.org/10.1063/1.451237>.
- [52] J.-M. Langlois, R.P. Muller, T.R. Coley, W.A. Goddard, M.N. Ringnalda, Y. Won, R.A. Friesner, Pseudospectral generalized valence-bond calculations: application to methylene, ethylene, and silylene, *J. Chem. Phys.* 92 (12) (1990) 7488–7497, <https://doi.org/10.1063/1.458184>.
- [53] M.N. Ringnalda, M. Belhadj, R.A. Friesner, Pseudospectral Hartree–Fock theory: applications and algorithmic improvements, *J. Chem. Phys.* 93 (5) (1990) 3397–3407, <https://doi.org/10.1063/1.458819>.
- [54] R.A. Friesner, New methods for electronic structure calculations on large molecules, *Annu. Rev. Phys. Chem.* 42 (1) (1991) 341–367, <https://doi.org/10.1146/annurev.pc.42.100191.002013>.
- [55] T.J. Martinez, A. Mehta, E.A. Carter, Pseudospectral full configuration interaction, *J. Chem. Phys.* 97 (3) (1992) 1876–1880, <https://doi.org/10.1063/1.463176>.
- [56] T.J. Martinez, E.A. Carter, Pseudospectral double excitation configuration interaction, *J. Chem. Phys.* 98 (9) (1993) 7081–7085, <https://doi.org/10.1063/1.464751>.
- [57] T.J. Martinez, E.A. Carter, Pseudospectral Møller–Plesset perturbation theory through third order, *J. Chem. Phys.* 100 (5) (1994) 3631–3638, <https://doi.org/10.1063/1.466350>.
- [58] T.J. Martinez, E.A. Carter, Pseudospectral multireference single and double excitation configuration interaction, *J. Chem. Phys.* 102 (19) (1995) 7564–7572, <https://doi.org/10.1063/1.469088>.
- [59] C. Ko, D.K. Malick, D.A. Braden, R.A. Friesner, T.J. Martínez, Pseudospectral time-dependent density functional theory, *J. Chem. Phys.* 128 (10) (Mar. 2008), <https://doi.org/10.1063/1.2834222>.
- [60] E.G. Hohenstein, R.M. Parrish, T.J. Martínez, Tensor hypercontraction density fitting. I. Quartic scaling second- and third-order Møller–Plesset perturbation theory, *J. Chem. Phys.* 137 (4) (2012) 044103, <https://doi.org/10.1063/1.4732310>.
- [61] R.M. Parrish, E.G. Hohenstein, T.J. Martínez, C.D. Sherrill, Tensor hypercontraction. ii. least-squares renormalization, *J. Chem. Phys.* 137 (22) (Dec. 2012), <https://doi.org/10.1063/1.4768233>.
- [62] K. Pierce, V. Rishi, E.F. Valeev, Robust approximation of tensor networks: application to grid-free tensor factorization of the Coulomb interaction, *J. Chem. Theory Comput.* 17 (4) (2021) 2217–2230, <https://doi.org/10.1021/acs.jctc.0c01310>.
- [63] L. Boehnke, H. Hafermann, M. Ferrero, F. Lechermann, O. Parcollet, Orthogonal polynomial representation of imaginary-time Green’s functions, *Phys. Rev. B* 84 (2011) 075145, <https://doi.org/10.1103/PhysRevB.84.075145>.
- [64] A.A. Kananenka, A.R. Welden, T.N. Lan, E. Gull, D. Zgid, Efficient temperature-dependent Green’s function methods for realistic systems: using cubic spline interpolation to approximate matsubara Green’s functions, *J. Chem. Theory Comput.* 12 (5) (2016) 2250–2259, <https://doi.org/10.1021/acs.jctc.6b00178>.
- [65] E. Gull, S. Isakov, I. Krivenko, A.A. Rusakov, D. Zgid, Chebyshev polynomial representation of imaginary-time response functions, *Phys. Rev. B* 98 (2018) 075127, <https://doi.org/10.1103/PhysRevB.98.075127>.
- [66] H. Shinaoka, J. Otsuki, M. Ohzeki, K. Yoshimi, Compressing Green’s function using intermediate representation between imaginary-time and real-frequency domains, *Phys. Rev. B* 96 (2017) 035147, <https://doi.org/10.1103/PhysRevB.96.035147>.
- [67] J. Kaye, K. Chen, O. Parcollet, Discrete Lehmann representation of imaginary time Green’s functions, *Phys. Rev. B* 105 (2022) 235115, <https://doi.org/10.1103/PhysRevB.105.235115>.
- [68] P. Pokhilko, C.-N. Yeh, D. Zgid, Iterative subspace algorithms for finite-temperature solution of Dyson equation, *J. Chem. Phys.* 156 (9) (2022) 094101, <https://doi.org/10.1063/5.0082586>.
- [69] Y. Yu, S. Isakov, E. Gull, Heating and cooling in self-consistent many-body simulations, *Phys. Rev. B* 108 (2023) 155116, <https://doi.org/10.1103/PhysRevB.108.155116>.
- [70] M. Jarrell, J. Gubernatis, Bayesian inference and the analytic continuation of imaginary-time quantum Monte Carlo data, *Phys. Rep.* 269 (3) (1996) 133–195, [https://doi.org/10.1016/0370-1573\(95\)00074-7](https://doi.org/10.1016/0370-1573(95)00074-7).
- [71] H.J. Vidberg, J.W. Serene, Solving the Eliashberg equations by means of N-point Padé approximants, *J. Low Temp. Phys.* 29 (3) (1977) 179–192, <https://doi.org/10.1007/BF00655090>.
- [72] J. Fei, C.-N. Yeh, E. Gull, Nevanlinna analytical continuation, *Phys. Rev. Lett.* 126 (2021) 056402, <https://doi.org/10.1103/PhysRevLett.126.056402>.
- [73] J. Fei, C.-N. Yeh, D. Zgid, E. Gull, Analytical continuation of matrix-valued functions: Carathéodory formalism, *Phys. Rev. B* 104 (2021) 165111, <https://doi.org/10.1103/PhysRevB.104.165111>.
- [74] Z. Huang, E. Gull, L. Lin, Robust analytic continuation of Green’s functions via projection, pole estimation, and semidefinite relaxation, *Phys. Rev. B* 107 (2023) 075151, <https://doi.org/10.1103/PhysRevB.107.075151>.
- [75] L. Ying, Analytic continuation from limited noisy matsubara data, *J. Comput. Phys.* 469 (2022) 111549, <https://doi.org/10.1016/j.jcp.2022.111549>.
- [76] L. Ying, Pole recovery from noisy data on imaginary axis, *J. Sci. Comput.* 92 (3) (Aug. 2022), <https://doi.org/10.1007/s10915-022-01963-z>.
- [77] L. Zhang, E. Gull, Minimal pole representation and controlled analytic continuation of matsubara response functions, *arXiv:2312.10576*, 2023.
- [78] A. Szabo, N. Ostlund, *Modern Quantum Chemistry: Introduction to Advanced Electronic Structure Theory*, Dover Books on Chemistry, Dover Publications, 1996, <https://books.google.com/books?id=k-DcGAAQBAJ>.
- [79] Q. Sun, X. Zhang, S. Banerjee, P. Bao, M. Barbry, N.S. Blunt, N.A. Bogdanov, G.H. Booth, J. Chen, Z.-H. Cui, J.J. Eriksen, Y. Gao, S. Guo, J. Hermann, M.R. Hermes, K. Koh, P. Koval, S. Lehtola, Z. Li, J. Liu, N. Mardirossian, J.D. McClain, M. Motta, B. Mussard, H.Q. Pham, A. Pulkin, W. Purwanto, P.J. Robinson, E. Ronca, E.R. Sayfutyarova, M. Scheurer, H.F. Schurkus, J.E.T. Smith, C. Sun, S.-N. Sun, S. Upadhyay, L.K. Wagner, X. Wang, A. White, J.D. Whitfield, M.J. Williamson, S. Wouters, J. Yang, J.M. Yu, T. Zhu, T.C. Berkelbach, S. Sharma, A.Y. Sokolov, G.K.-L. Chan, Recent developments in the PySCF program package, *J. Chem. Phys.* 153 (2) (2020) 024109, <https://doi.org/10.1063/5.0006074>.
- [80] R. Mattuck, *A Guide to Feynman Diagrams in the Many-Body Problem*, Dover Books on Physics Series, Dover Publications, 1992, <https://books.google.com/books?id=pe-v8zfxE68C>.
- [81] M. Born, *Quantenmechanik der stoßvorgänge*, *Z. Phys.* 38 (11) (1926) 803–827, <https://doi.org/10.1007/BF01397184>.
- [82] J.J. Phillips, D. Zgid, Communication: the description of strong correlation within self-consistent Green’s function second-order perturbation theory, *J. Chem. Phys.* 140 (24) (2014) 241101, <https://doi.org/10.1063/1.4884951>.
- [83] A.A. Rusakov, D. Zgid, Self-consistent second-order Green’s function perturbation theory for periodic systems, *J. Chem. Phys.* 144 (5) (2016) 054106, <https://doi.org/10.1063/1.4940900>.
- [84] S. Isakov, A.A. Rusakov, D. Zgid, E. Gull, Effect of propagator renormalization on the band gap of insulating solids, *Phys. Rev. B* 100 (2019) 085112, <https://doi.org/10.1103/PhysRevB.100.085112>.

- [85] M. Shishkin, M. Marsman, G. Kresse, Accurate quasiparticle spectra from self-consistent gw calculations with vertex corrections, *Phys. Rev. Lett.* 99 (2007) 246403, <https://doi.org/10.1103/PhysRevLett.99.246403>.
- [86] C.-N. Yeh, S. Iskakov, D. Zgid, E. Gull, Fully self-consistent finite-temperature gw in Gaussian Bloch orbitals for solids, *Phys. Rev. B* 106 (2022) 235104, <https://doi.org/10.1103/PhysRevB.106.235104>.
- [87] A. Kutevov, Self-consistent gw method: O(n) algorithm for polarizability and self energy, *Comput. Phys. Commun.* 257 (2020) 107502, <https://doi.org/10.1016/j.cpc.2020.107502>.
- [88] T. Saue, Relativistic Hamiltonians for chemistry: a primer, *ChemPhysChem* 12 (17) (2011) 3077–3094, <https://doi.org/10.1002/cphc.201100682>.
- [89] C.-N. Yeh, A. Shee, Q. Sun, E. Gull, D. Zgid, Relativistic self-consistent gw: exact two-component formalism with one-electron approximation for solids, *Phys. Rev. B* 106 (2022) 085121, <https://doi.org/10.1103/PhysRevB.106.085121>.
- [90] H.J. Monkhorst, J.D. Pack, Special points for Brillouin-zone integrations, *Phys. Rev. B* 13 (1976) 5188–5192, <https://doi.org/10.1103/PhysRevB.13.5188>.
- [91] N. Marzari, A.A. Mostofi, J.R. Yates, I. Souza, D. Vanderbilt, Maximally localized Wannier functions: theory and applications, *Rev. Mod. Phys.* 84 (2012) 1419–1475, <https://doi.org/10.1103/RevModPhys.84.1419>.
- [92] S. Fuchs, T. Pruschke, M. Jarrell, Analytic continuation of quantum Monte Carlo data by stochastic analytical inference, *Phys. Rev. E* 81 (2010) 056701, <https://doi.org/10.1103/PhysRevE.81.056701>.
- [93] K. Nogaki, J. Fei, E. Gull, H. Shinaoka, Nevanlinna.jl: a Julia implementation of Nevanlinna analytic continuation, *SciPost Phys. Codebases* (2023) 19, <https://doi.org/10.21468/SciPostPhysCodeB.19>.
- [94] I. Sergei, A. Hampel, N. Wentzell, E. Gull, Triqs/Nevalinna: implementation of the Nevanlinna analytic continuation method for noise-free data, <https://doi.org/10.48550/ARXIV.2309.01407>, 2023.
- [95] G. Guennebaud, B. Jacob, et al., Eigen v3, <http://eigen.tuxfamily.org>, 2010.
- [96] Message Passing Interface Forum, MPI: a message-passing interface standard version 4.0, <https://www.mpi-forum.org/docs/mpi-4.0/mpi40-report.pdf>, Jun. 2021.
- [97] The HDF Group, Hierarchical data format, version 5, <https://github.com/HDFGroup/hdf5>, 2024.
- [98] C.L. Lawson, R.J. Hanson, D.R. Kincaid, F.T. Krogh, Basic linear algebra subprograms for fortran usage, *ACM Trans. Math. Softw.* 5 (3) (1979) 308–323, <https://doi.org/10.1145/355841.355847>.
- [99] cmake, CMake is a tool to manage building of source code, <https://cmake.org/>, 2024.
- [100] cmake, GNU make is a tool which controls the generation of executables and other non-source files of a program from the program's source files, <https://www.gnu.org/software/make/>, 2024.
- [101] Ninja, Ninja is a small build system with a focus on speed, <https://ninja-build.org/>, 2024.
- [102] J. Nickolls, I. Buck, M. Garland, K. Skadron, Scalable parallel programming with cuda: is cuda the parallel programming model that application developers have been waiting for?, *Queue* 6 (2) (2008) 40–53.
- [103] T. Granlund, the GMP development team, GNU MP: The GNU Multiple Precision Arithmetic Library, 5th edition, 2012, <http://gmplib.org/>.
- [104] S.K. Lam, A. Pitrou, S. Seibert, Numba: a llvm-based python jit compiler, in: *Proceedings of the Second Workshop on the LLVM Compiler Infrastructure in HPC, LLVM '15*, Association for Computing Machinery, New York, NY, USA, 2015.
- [105] A. Togo, K. Shinohara, I. Tanaka, Spglib: a software library for crystal symmetry search, [arXiv:1808.01590](https://arxiv.org/abs/1808.01590), 2024.
- [106] A.H. Larsen, J.J. Mortensen, J. Blomqvist, I.E. Castelli, R. Christensen, M. Dulak, J. Friis, M.N. Groves, B. Hammer, C. Hargus, E.D. Hermes, P.C. Jennings, P.B. Jensen, J. Kermode, J.R. Kitchin, E.L. Kolsbjerg, J. Kubal, K. Kaasbjerg, S. Lysgaard, J.B. Maronsson, T. Maxson, T. Olsen, L. Pastewka, A. Peterson, C. Rostgaard, J. Schiøtz, O. Schütt, M. Strange, K.S. Thygesen, T. Vegge, L. Vilhelmsen, M. Walter, Z. Zeng, K.W. Jacobsen, The atomic simulation environment—a python library for working with atoms, *J. Phys. Condens. Matter* 29 (27) (2017) 273002, <http://stacks.iop.org/0953-8984/29/i=27/a=273002>.
- [107] PySCF built-in molecular basis sets, [link]. URL <https://github.com/pyscf/pyscf/tree/master/pyscf/gto/basis>.
- [108] PySCF built-in basis sets for periodic systems, [link]. URL <https://github.com/pyscf/pyscf/tree/master/pyscf/pbc/gto/basis>.
- [109] E. Aprà, E.J. Bylaska, W.A. de Jong, N. Govind, K. Kowalski, T.P. Straatsma, M. Valiev, H.J.J. van Dam, Y. Alexeev, J. Anchell, V. Anisimov, F.W. Aquino, R. Atta-Fynn, J. Autschbach, N.P. Bauman, J.C. Becca, D.E. Bernholdt, K. Bhaskaran-Nair, S. Bogatko, P. Borowski, J. Boschen, J. Brabec, A. Bruner, E. Cauët, Y. Chen, G.N. Chuev, C.J. Cramer, J. Daily, M.J.O. Deegan, T.H. Dunning Jr., M. Dupuis, K.G. Dyall, G.I. Fann, S.A. Fischer, A. Fonari, H. Früchtl, L. Gagliardi, J. Garza, N. Gawande, S. Ghosh, K. Glaesemann, A.W. Götz, J. Hammond, V. Helms, E.D. Hermes, K. Hirao, S. Hirata, M. Jacquelin, L. Jensen, B.G. Johnson, H. Jónsson, R.A. Kendall, M. Klemm, R. Kobayashi, V. Konkov, S. Krishnamoorthy, M. Krishnan, Z. Lin, R.D. Lins, R.J. Littlefield, A.J. Logsdail, K. Lopata, W. Ma, A.V. Marenich, J. Martin del Campo, D. Mejia-Rodriguez, J.E. Moore, J.M. Mullin, T. Nakajima, D.R. Nascimento, J.A. Nichols, P.J. Nichols, J. Nieplocha, A. Otero-de-la Roza, B. Palmer, A. Panyala, T. Pirojsirikul, B. Peng, R. Peverati, J. Pittner, L. Pollack, R.M. Richard, P. Sadayappan, G.C. Schatz, W.A. Shelton, D.W. Silverstein, D.M.A. Smith, T.A. Soares, D. Song, M. Swart, H.L. Taylor, G.S. Thomas, V. Tipparaju, D.G. Truhlar, K. Tsemekhman, T. Van Voorhis, A. Vazquez-Mayagoitia, P. Verma, O. Villa, A. Vishnu, K.D. Vogiatzis, D. Wang, J.H. Weare, M.J. Williamson, T.L. Windus, K. Woliński, A.T. Wong, Q. Wu, C. Yang, Q. Yu, M. Zacharias, Z. Zhang, Y. Zhao, R.J. Harrison, NWChem: past, present, and future, *J. Chem. Phys.* 152 (18) (2020) 184102, <https://doi.org/10.1063/5.0004997>.
- [110] PySCF built-in pseudopotentials for periodic systems, [link]. URL <https://github.com/pyscf/pyscf/tree/master/pyscf/pbc/gto/pseudo>.
- [111] [link]. URL https://molssi-bse.github.io/basis_set_exchange/conversion.html.
- [112] F. Gygi, A. Baldereschi, Self-consistent Hartree-Fock and screened-exchange calculations in solids: application to silicon, *Phys. Rev. B* 34 (1986) 4405–4408, <https://doi.org/10.1103/PhysRevB.34.4405>.
- [113] J. Paier, R. Hirschl, M. Marsman, G. Kresse, The Perdew–Burke–Ernzerhof exchange-correlation functional applied to the G2-1 test set using a plane-wave basis set, *J. Chem. Phys.* 122 (23) (2005) 234102, <https://doi.org/10.1063/1.1926272>.
- [114] P. Broqvist, A. Alkauskas, A. Pasquarello, Hybrid-functional calculations with plane-wave basis sets: effect of singularity correction on total energies, energy eigenvalues, and defect energy levels, *Phys. Rev. B* 80 (2009) 085114, <https://doi.org/10.1103/PhysRevB.80.085114>.
- [115] H. Fukutome, Unrestricted Hartree–Fock theory and its applications to molecules and chemical reactions, *Int. J. Quant. Chem.* 20 (5) (1981) 955–1065, <https://doi.org/10.1002/qua.560200502>.
- [116] P. Pokhilko, D. Zgid, Broken-symmetry self-consistent GW approach: degree of spin contamination and evaluation of effective exchange couplings in solid antiferromagnets, *J. Chem. Phys.* 157 (14) (2022) 144101, <https://doi.org/10.1063/5.0114080>.
- [117] P. Pokhilko, D. Zgid, Evaluation of neel temperatures from fully self-consistent broken-symmetry gw and high-temperature expansion: application to cubic transition-metal oxides, *J. Phys. Chem. Lett.* 14 (25) (2023) 5777–5783, <https://doi.org/10.1021/acs.jpclett.3c00760>, PMID: 37326965.
- [118] P. Pulay, Convergence acceleration of iterative sequences. The case of scf iteration, *Chem. Phys. Lett.* 73 (2) (1980) 393–398, [https://doi.org/10.1016/0009-2614\(80\)80396-4](https://doi.org/10.1016/0009-2614(80)80396-4).
- [119] P. Pulay, Improved scf convergence acceleration, *J. Comput. Chem.* 3 (4) (1982) 556–560, <https://doi.org/10.1002/jcc.540030413>.
- [120] J. VandeVondele, J. Hutter, Gaussian basis sets for accurate calculations on molecular systems in gas and condensed phases, *J. Chem. Phys.* 127 (11) (2007) 114105, <https://doi.org/10.1063/1.2770708>.
- [121] J. Li, M. Wallerberger, N. Chikano, C.-N. Yeh, E. Gull, H. Shinaoka, Sparse sampling approach to efficient ab initio calculations at finite temperature, *Phys. Rev. B* 101 (2020) 035144, <https://doi.org/10.1103/PhysRevB.101.035144>.
- [122] M. Wen, V. Abraham, G. Harsha, A. Shee, K.B. Whaley, D. Zgid, Comparing self-consistent gw and vertex-corrected g0w0 (g0w0 γ) accuracy for molecular ionization potentials, *J. Chem. Theory Comput.* 20 (8) (2024) 3109–3120, <https://doi.org/10.1021/acs.jctc.3c01279>.
- [123] V. Abraham, G. Harsha, D. Zgid, Relativistic fully self-consistent gw for molecules: total energies and ionization potentials, *J. Chem. Theory Comput.* 20 (11) (2024) 4579–4590, <https://doi.org/10.1021/acs.jctc.4c00075>.