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The Generalized Green's function Cluster Expansion: A Python package for simulating polarons

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Software

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Summary

We present an efficient implementation of the Generalized Green's function Cluster Expansion (GGCE), which is a new method for computing the ground-state properties and dynamics of polarons (single electrons coupled to lattice vibrations) in model electron-phonon systems. The GGCE works at arbitrary temperature and is well suited for a variety of electron-phonon couplings, including, but not limited to, site and bond Holstein and Peierls (Su-Schrieffer-Heeger) couplings, and couplings to multiple phonon modes with different energy scales and coupling strengths. Quick calculations can be performed efficiently on a laptop using solvers from NumPy and SciPy, or in parallel at scale using the PETSc sparse linear solver engine.

Statement of need

The electron-phonon problem is of both fundamental relevance and practical importance in materials science (Mahan, 2010; Peter & Cardona, 2010). Electron-phonon interactions promote a variety of states, low-temperature phases and high-temperature transport phenomena in quantum materials. For example, they are essential to the understanding of the behavior of solar cells (Schilcher et al., 2021) and semiconductors (Fratini et al., 2020). In the dilute-carrier-density limit, electron-phonon coupling gives rise to quasiparticles called polarons whose properties encode the physics of materials in various temperature regimes.

Research on polarons has been divided into two thrusts: fundamental theoretical work focused on qualitative physical aspects (Mahan, 2010) and applied research focused on obtaining quantitative properties relevant to specific materials (Giustino, 2017; Lee et al., 2018; Poncé et al., 2016; Sio et al., 2019b, 2019a; Zhou et al., 2021; Zhou & Bernardi, 2016). This paper presents a new scientific software that aims to bridge this gap. It allows for the treatment of polaron statics and dynamics in models of electron-phonon coupling of almost arbitrary form provided that they are sufficiently short-ranged. It presents a self-contained first step in an ongoing effort to combine an *ab initio* understanding of materials and exact many-body analysis of polaron states.

Software summary

The GGCE method is a *numerically exact* extension of a family of variational approaches known in the theoretical physics community as Momentum Average (MA) methods (Berciu, 2006; Glen L. Goodvin et al., 2006). Details on the theoretical framework of GGCE can be found in Carbone *et al* (Carbone, Reichman, et al., 2021; Carbone, Millis, et al., 2021). Our code, named for the method, is a Python package meant to make implementing the GGCE framework as straightforward as possible. In addition, through only slight modifications to our standard API, the user can invoke powerful PETSc sparse solvers for massively parallel computations at scale (Balay et al., 1997, 2022a, 2022b; Zhang et al., 2022).

A fundamental insight of the MA approximation is to utilize a variational space formed of clouds of spatially clustered phonon configurations with the electron allowed to be anywhere in the system. Through comparison with exact methods, this approximation was shown to yield quantitatively accurate results. In order to systematically converge MA to the limit of infinite Hilbert space dimension, the cloud size and total phonon number, which serve as control parameters, are taken to infinity (Glen L. Goodvin et al., 2006). This, however, requires the derivation of a set of equations corresponding to a given cloud size for all cloud sizes smaller than a cutoff. This cutoff is then increased until convergence is achieved. The ever-increasing complexity of the structure of the system of equations at large cloud sizes means that this approach can very quickly become intractable to do by hand (Marchand et al., 2010; Sous et al., 2017, 2018), especially in the regime of small phonon energies where large cloud sizes are usually needed in order to converge to the numerically exact limit. Carbone *et al* proposed a generalized implementation of the MA method which automates the generation and solution of the systems of equations for arbitrary cloud sizes (Carbone, Reichman, et al., 2021). Benchmarks of GGCE on several model systems verified that convergence with cloud size is fast, rendering this an efficient and controlled numerically exact method even in challenging parameter regimes.

Previous studies using GGCE focused on polarons at zero temperature. We also include a new functionality which allows the study of polarons at finite temperature. Here, we make use of the Thermofield Double formalism (Suzuki, 1985; Takahashi & Umezawa, 1996), which exactly maps any given model at finite temperature to one at zero temperature with couplings to real and fictitious phonons. This model can be solved naturally using the apparatus of the zero-temperature GGCE method. Benchmarks of this approach are ongoing, and preliminary results suggest that the method may be competitive with state-of-the-art methods. A paper with these results is currently in preparation.

Formally, the GGCE functions as an on-the-fly generator of equations of motion for the single-particle Green's function given a set of control parameters (cloud size, total phonon number, and their extensions to systems with multiple phonon modes) and input model parameters (energy scales, coupling strength, etc.). The generated system of equations is then solved numerically in order to obtain the Green's function of interest using a chosen solver. Furthermore, the equation of motion dictates how different Green's functions or propagators couple, and so one can use the solver to numerically obtain any one particular propagator or a set of them, which can be used to construct other quantities such as the optical conductivity (Glen L. Goodvin et al., 2011) or resonant inelastic X-ray scattering spectrum (Bieniasz et al., 2021).

The GGCE code consists of three components detailed in our documentation: models, systems and solvers.

- Models completely describe the Hamiltonian system to solve, and the level of theory (specified by the control parameters) at which to solve it;
- Systems construct all of the objects required to build the matrix to solve the system of equations;
- Solvers utilize different back-ends to actually solve the constructed matrix in an efficient

manner.

Models

The choice of model completely defines the type of electron-phonon coupling used in the Hamiltonian. Every model Hamiltonian implemented so far assumes a lattice with nearest-neighbor hopping of electrons and Einstein (dispersionless) phonons. The user can set the electron hopping, phonon frequency and type and strength of the electron-phonon coupling. Currently, we have implemented the Holstein, site Peierls (site Su-Schrieffer-Heeger), bond Peierls (bond Su-Schrieffer-Heeger) and Edwards Fermion-boson models (as well as any arbitrary combination of these).

Systems

The Systems objects are a helpful intermediary for performing the sometimes expensive step of constructing the Python objects required for building matrices of linear equations. Systems are instantiated from a Model. At creation, using the information in the Model about the electron-phonon couplings, Systems automatically construct and store the equations-of-motion object called the "basis". The basis remains "un-evaluated": it is passed into the Solver, where it can be used to obtain a matrix of equation coefficients at any values of momentum and frequency. In this way, the basis is constructed only once, and then simply called repeatedly to determine the coefficients. Construction of the basis follows the scheme outlined in Carbone *et al* (Carbone, Reichman, et al., 2021). For relatively small clouds, the basis can be visualized for sanity-checking the calculation, using a method that pretty-prints the equations' structure.

When provided a directory, the System will also automatically checkpoint the basis to disk using `pickle`, allowing for a later restart of a failed computation or for restarting long jobs on a cluster with time-limited jobs without the expensive re-computation of the basis.

Solvers

At the heart of GGCE are the Solvers, which implement different approaches to solving the linear systems of equations obtained by a System object. The simplest of these uses NumPy's dense solver, which solves the equation-of-motion matrix using an efficient continued fraction approach (Glen L. Goodvin et al., 2006), or SciPy's sparse solver. For truly large-scale computations with sizable phonon clouds and/or many different electron-phonon couplings operating simultaneously, GGCE interfaces with the powerful, massively parallel PETSc sparse solver engine. All GGCE Solvers are MPI-enabled, and allow for a variety of parallelization schemes, all of which are detailed in our documentation. At the extreme, the PETSc interface can parallelize calculations across momentum-frequency points and also parallelize the solving of a single large sparse matrix at each point, and thus allowing for straightforward use of all available computational resources.

The Solver allows the user to quickly evaluate the Green's function for a specified range of momenta and frequencies. Like the System, it automatically checkpoints the solution (Green's function value) at every momentum-frequency point using `pickle`, allowing for restart in case of failure or time limits on cluster jobs.

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