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PAOFLOW: A utility to construct and operate on *ab initio* Hamiltonians from the projections of electronic wavefunctions on atomic orbital bases, including characterization of topological materials

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ABSTRACT

PAOFLOW is a utility for the analysis and characterization of materials properties from the output of electronic structure calculations. By exploiting an efficient procedure to project the full plane-wave solution on a reduced space of atomic orbitals, PAOFLOW facilitates the calculation of a plethora of quantities such as diffusive, anomalous and spin Hall conductivities, magnetic and spin circular dichroism, and Z_2 topological invariants and more. The computational cost associated with post-processing first principles calculations is negligible. This code, written entirely in Python under GPL 3.0 or later, opens the way to the high-throughput computational characterization of materials at an unprecedented scale.

1. Introduction

PAOFLOW is a software tool to efficiently post-process standard first principles electronic structure plane-wave pseudopotential calculations in order to promptly compute from interpolated band structures and density of states several quantities that provide insight on transport, optical, magnetic and topological properties such as anomalous and spin Hall conductivity (AHC and SHC, respectively), magnetic circular dichroism, spin circular dichroism, and topological invariants. The methodology is based on the projection on pseudo-atomic orbitals (PAO) [1–3] and is the latest addition to the AFLOW software infrastructure [4,5]. Additional features of PAOFLOW include the calculation of selected integrated quantities using adaptive smearing, the ability to add spin orbit coupling using parametrized methods, and the calculation of surface projected band structures.

PAOFLOW is massively parallel by design (both CPU and GPU) and provides the user with the ability to determine measurable quantities with first principles accuracy and with the speed and robustness required by high-throughput materials characterization. The current implementation (using Quantum ESPRESSO, QE) [6,7] does not require any additional input with respect to a standard electronic structure calculations and, seamlessly, provides a real space tight-binding (TB) representation of the Hamiltonian matrix in a self-contained XML format. The sparse PAO matrix can be easily Fourier transformed and interpolated to determine the full energy dispersion and to compute additional properties associated with derivatives of the energy bands, such as matrix elements of the momentum operator or electron velocities, with the desired level of resolution. PAOFLOW is publicly available under the terms of the GNU General Public License as published by the Free Software Foundation, either version 3 of the License, or any later version. PAOFLOW is integrated in the AFLOWπ high-throughput framework [8] and it is distributed at http://www.aflow.org/src/aflowpi and http://www.aflow.org/src/paoflow.

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2. Software design

PAOFLOW is written in Python 2.7 (using the Python standard libraries `NumPy` and `SciPy`). The systematic use of regular expression (`re` module) and XML parsing (`xml.etree.cElementTree` module) makes the software expandable to a variety of electronic structure engines with minimal effort. Parallelization on CPUs uses the openMPI protocol through the `mpi4py` module, while GPU parallelization is based on the CUDA kernel through the `pycuda`[1] and `skcuda` [9] modules.

Currently, PAOFLOW requires few basic calculations with the QE package: a first one to generate converged electronic density and Kohn–Sham (KS) potential on an appropriate Monkhorst and Pack (MP) **k**-point mesh (`pw.x`), a second non self consistent calculation (`pw.x`) to evaluate eigenvalues and eigenfunctions for a MP mesh centered at Γ (**k** = (0,0,0), `nosym` and `noinv` = `.true.`) and a third post-processing run using `projwfc.x` to obtain the projection of the eigenfunctions on the pseudo atomic basis functions. No additional calculations with QE are required.

Starting with highly interpolated first principles electronic properties (Fig. 1), PAOFLOW computes band derivatives and Berry's curvature (Fig. 2). These ingredients are then used to determine efficiently the AHC (Fig. 3), magnetic circular dichroism spectra (Fig. 4), SHC (Fig. 5), and spin circular dichroism (Fig. 6).

The PAOFLOW package is distributed with several examples (in the main directory of the distribution, see Section 6) describing the computable physical quantities and can be easily installed on any hardware.

3. Description of the code

– Modules: `build_Pn.py`, `build_Hks.py`
Accurate PAO Hamiltonian matrices can be built from the direct projection of the KS Bloch states $|\psi_{n\mathbf{k}}\rangle$ onto a chosen basis set of fixed localized functions, as it was discussed extensively in Ref. [1–3]. The Hamiltonian for a specific material, $\widehat{H}(\mathbf{r}_\alpha)$, is computed in real space using atomic orbitals or pseudo atomic orbitals from the pseudopotential of any given element. The key in this procedure is in the mapping of the *ab initio* electronic structure (solved on a well converged and large plane waves basis set) into a model that precisely reproduces a selected number of bands of interest. The crucial quantities that measure the accuracy of the basis set are the projectabilities $p_{n\mathbf{k}} = \langle\psi_{n\mathbf{k}}|\widehat{P}|\psi_{n\mathbf{k}}\rangle \geqslant 0$ ($\widehat{P}$ is the operator that projects onto the space of the PAO basis set, as defined in Ref. [2]) which indicate the representability of a Bloch state $|\psi_{n\mathbf{k}}\rangle$ on the chosen PAO set. Maximum projectability, $p_{n\mathbf{k}} = 1$, indicates that the particular Bloch state can be perfectly represented in the chosen PAO set; contrarily, $p_{n\mathbf{k}} \approx 0$ indicates that the PAO set is insufficient and should be augmented. Once the Bloch states with good projectabilities have been identified, the PAO Hamiltonian is constructed either as

$$\widehat{H}(\mathbf{k}) = AEA^\dagger + \kappa\left(I - AA^\dagger\right) \tag{1}$$

following Ref. [1] or

$$\widehat{H}(\mathbf{k}) = AEA^\dagger + \kappa\left(I - A\left(A^\dagger A\right)^{-1}A^\dagger\right) \tag{2}$$

as in Ref. [2], where the case can be chosen in the input of PAOFLOW (see Listing 2 below). Here E is the diagonal matrix of KS eigenenergies and A is the matrix of coefficients obtained from projecting the Bloch wavefunctions onto the PAO set. Since the filtering procedure introduces a null space, the parameter κ is used to shift all the unphysical solutions outside a given energy range of interest. The procedure in Eq. (2) is recommended for most cases. This procedure provides an accurate real space representation of the *ab initio* Hamiltonian $\widehat{H}(\mathbf{R})$ as a TB matrix of small dimension written in XML format, a crucial advantage for the accurate calculation of any physical properties that requires the precise integration in the reciprocal space.

– Module: `add_ext_field.py`
Following the TB formalism (see, for instance, Ref. [15]), the user can add an arbitrary external electric field as a (time-dependent) scalar potential acting on the diagonal elements of

[1] https://mathema.tician.de/software/pycuda/.

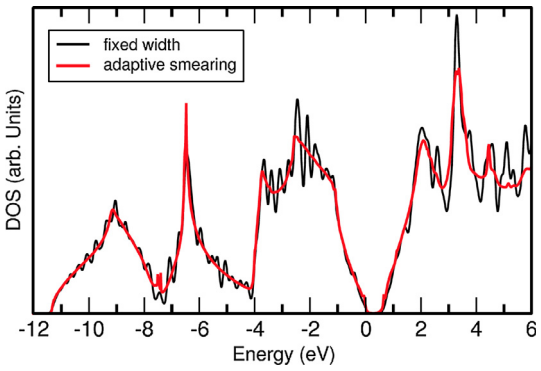

Fig. 1. Comparison between fixed width and adaptive smearing for the density of state of FCC Si. The calculation is done on a 18 × 18 × 18 MP grid and with W = 0.1 eV in the fixed width smearing algorithm. Data adapted from `example 01`.

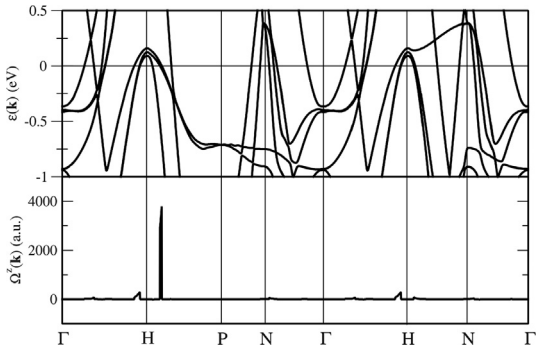

Fig. 2. Top panel: band structure of Fe including spin-orbit interaction. Bottom panel: berry curvature. Data adapted from `example 04` and evaluated along the AFLOW standard path for the BCC lattice [10].

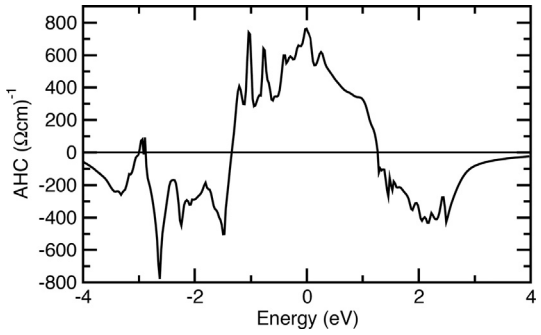

Fig. 3. Energy resolved anomalous Hall conductivity in Fe. Data adapted from `example 04` using a 42 × 42 × 42 MP grid and adaptive broadening yielding a converged value for the AHC at E_{Fermi} of 751 $(\Omega\text{cm})^{-1}$ in excellent agreement with the results from Ref. [11].