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FHI-vibes: *Ab Initio* Vibrational Simulations

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Introduction

The vibrational motion of nuclei determines many important properties of materials, including their thermodynamic equilibrium and non-equilibrium properties. Accurately assessing the nuclear dynamics and the associated material properties is therefore an important task for computational materials scientists in a broad range of sub-fields. Of particular importance are simulation techniques that build on first-principles electronic-structure simulations and thereby allow to systematically investigate the virtually infinite space of materials, including those systems for which little or no experimental data is hitherto available (Curtarolo et al., 2013). This allows one to design novel and improved materials with optimal properties for many applications, e.g., high-performance thermal insulators for gas and airplane turbines (Evans et al., 2008), organic semicondcutors with long-term phase stabilities (Salzillo et al., 2016), thermoelectric generators (Snyder & Toberer, 2008), and improved thermal management systems (Tian & Ren, 2019).

Essentially, there are two distinct routes towards assessing vibrational properties: In perturbative *lattice dynamics* techniques, the potential-energy surface on which the nuclei move is approximated with a Taylor expansion around the equilibrium structure. Typically, one starts from a second-order expansion, i.e., the *harmonic approximation*, which allows for an analytic solution of the equations of motion (Dove, 1993) and thus for a straightforward evaluation of observables (thermodynamic expectation values). Higher-order terms in the Taylor expansion can be accounted for perturbatively. Conversely, *molecular dynamics* (MD) based approaches account for the full, non-perturbative potential-energy surface *without* approximating the actual interactions. This requires one to solve the equations of motion numerically by propagating the atoms in time; physical properties can then be extracted as time averages of properly chosen observables (Tuckerman, 2010). Although both *lattice dynamics* and *molecular dynamics* techniques aim at computing the same physical observables, the involved methodologies, formalisms, and challenges are quite different. Accordingly, both methodologies also have different strengths and weaknesses: For instance, performing and analyzing MD simulations is typically computationally and conceptually more challenging, whereas perturbative lattice dynamics calculations inherently rely on approximations that are hard to validate.

To date, a variety of different software packages exists at different degrees of sophistication in both fields. Prominent examples are the *phonopy* code (Togo & Tanaka, 2015) for performing *lattice dynamics* calculations using Parlinski's finite-difference formalism (Parlinski et al., 1997) and the *i-PI* code (Kapil et al., 2019) for performing classical MD and quantum-mechanical path-integral MD simulations. Both packages interface with a variety of first-principles codes like *VASP* (Kresse & Furthmüller, 1996), *QuantumEspresso* (Giannozzi et al., 2009), *Abinit* (Gonze et al., 2020), *FHI-aims* (Blum et al., 2009), and several others.

Statement of need

To date, there is no software solution that allows for the seamless bridging and interlinking of *lattice dynamics* and *molecular dynamics* based approaches, despite the fact that actual material science studies can profit in accuracy and efficiency by exploiting both approaches. For instance, potential use cases include accelerating *MD* calculations by starting from harmonic equilibrium configurations (West & Estreicher, 2006), analyzing *MD* simulations in terms of harmonic phonons (Turney et al., 2009), investigating the range of validity of the perturbative expansion used in *lattice dynamics* (Knoop et al., 2020), and overcoming finite-size and finite-time effects in *ab initio* Green Kubo simulations of the thermal conductivity (Carbogno et al., 2017). Given the wide opportunities for application, the aspect of *integration*, i.e., the ability to utilize different methodologies from distinct codes in a flexible fashion using a consistent user interface, is paramount. In particular, this is a prerequisite for automatizing these workflows to enable hierarchical high-throughput screening of whole material classes in a systematic fashion. For example, such a workflow would start from geometry optimizations followed by a study of harmonic properties for many materials, so to single out candidate materials for more involved, fully anharmonic aiMD simulation techniques. Along these lines, let us mention that providing descriptive input and output files is a prerequisite for sharing raw data and results in a transparent and interpretable way in the spirit of open science and the FAIR Principles (Draxl & Scheffler, 2018). On top of that, tracking the provenance (Huber et al., 2020) across different codes facilitates the repurposing and analysis of the obtained data.

Summary

FHI-vibes is a *python* package that allows for such an integrated workflow. It uses the *Atomistic Simulation Environment (ASE)* (Hjorth Larsen et al., 2017) as a backend in order to represent materials and to connect to various first-principles codes. Via *ASE*, *FHI-vibes* provides a flexible framework for geometry optimization and MD, and connects to external codes like *spglib* (Togo & Tanaka, 2018), *phonopy* (Togo & Tanaka, 2015), *phono3py* (Togo et al., 2015), and *hiphive* (Eriksson et al., 2019) that implement lattice dynamics techniques based on the harmonic approximation. For all these tasks, *FHI-vibes* provides defined input files and a command line interface to set up and run calculations on local machines and clusters using the *slurm* submission system. The output is organized in self-contained and descriptive output files that enable a straightforward exchange of the data obtained with different methodologies. For advanced analysis, it provides an API fully compatible with *ASE* as well as *numpy* (van der Walt et al., 2011), *pandas* (McKinney, 2011), and *xarray* (Hoyer & Hamman, 2017); several user-friendly utilities allow to perform the most common postprocessing tasks within the command-line interface, such as providing comprehensive summaries of MD simulations or phonon calculations.

FHI-vibes provides a connection to *FireWorks* (Jain et al., 2015), a workflow management system for running simulation workflows on extensive sets of materials in high-throughput fashion. *FHI-vibes* is tightly integrated with *FHI-aims* (Blum et al., 2009) to perform energy and force calculations, but extending the functionality to any calculator available via *ASE* is straightforward.

FHI-vibes was used to produce the results in (Knoop et al., 2020).

Features

To facilitate the scientific studies described in the statement of need, *FHI-vibes 1.0* offers the following main features:

- Free and symmetry-constrained geometry optimization,
- harmonic phonon calculations,
- molecular dynamics simulations,
- harmonic sampling, and
- anharmonicity quantification.

An extensive user guide including tutorials and a reference documentation for these features is available at `vibes.fhi-berlin.mpg.de`. As demonstrated in a dedicated tutorial, the tasks can be easily combined and tailored to define workflows for high-throughput screening of material space.

The codebase and user interface of *FHI-vibes* are designed as a modular framework such that more advanced features and workflows are straightforward to add in the future.

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