Vibrant: A Post-Processing Tool for Computational Vibrational Spectroscopy of Molecules, Liquids and Materials

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Summary

In this work, we present Vibrant, a vibrational analysis program written in Fortran 2008. It interfaces to the FFTW (Frigo & Johnson, 1998) and GreenX (GX-AC) (Leucke et al., 2025) libraries and is parallelized using OpenMP for computational efficiency. Vibrant enables the generation of vibrational spectra for gases, liquids and materials through post-processing atomic positions, velocities, forces, dipole moments and polarizability tensors. Its functionalities include the computation of vibrational frequencies, as well as infrared (IR) and Raman intensities, using either static or molecular dynamics (MD)-based approaches. In addition, Vibrant enables the computation of resonance Raman (RR) and absorption spectra based on real-time propagation trajectories. Vibrant is distributed under the Apache 2.0 license and is available as open-source software on GitHub.

Statement of need

Vibrational spectroscopy, particularly IR and Raman techniques, provides valuable insight into the structure of gaseous, liquid, and solid materials. While in conventional Raman spectroscopy the excitation is non-resonant with electronic transitions, RR spectroscopy occurs when the excitation wavelength matches such a transition (Long, 2002). The resulting enhancement of specific vibrational modes allows, for example, *in situ* monitoring of reactants or products through characteristic marker bands during synthesis (Reichmayr et al., 2025).

Computational simulation of vibrational spectroscopy is often not straightforward. Most quantum chemistry packages that perform electronic structure calculations provide only the dipole moments or polarizability tensors, rather than the final spectrum itself. The challenge becomes greater when one goes beyond the static regime to compute vibrational spectra, such as when performing MD simulations to capture anharmonic modes or solvent effects, or real-time time-dependent density functional theory (RT-TDDFT) simulations to access excited state dynamics, relevant for RR spectroscopy.

Post-processing programs that can handle these properties are often limited in scope and scattered across different software packages. In this work, we introduce Vibrant, a computational tool which bridges this gap by performing a series of post-processing procedures on dipole moment and polarizability data to generate MD-based or static vibrational spectra. Our code can provide these spectra for gases, liquids and a wide range of materials. Figure 1 provides an overview of Vibrant's major functionalities.

The first panel (top-left) summarizes Vibrant's frequency analysis capabilities, which include

⁴¹ static calculations within the harmonic approximation and post-processing of MD trajectories. In
⁴² the static approach, conventional normal mode analysis is used to obtain vibrational frequencies
⁴³ and normal mode coordinates, while in the MD-based approach, normal mode derivatives
⁴⁴ are replaced by Fourier transforms of time-autocorrelation functions, yielding power spectra
⁴⁵ (Gordon, 1965; Thomas et al., 2013).

⁴⁶ The second panel (top-right) highlights Vibrant's functionality for computing IR, non-resonant
⁴⁷ Raman and RR spectral intensities, which can be obtained from either static or MD-based
⁴⁸ calculations. For IR and non-resonant Raman spectroscopy, Vibrant can handle different types
⁴⁹ of dipole moments and polarizabilities. Dipole moments may be provided as Berry-phase dipoles
⁵⁰ or generated from Wannier centers, while polarizabilities can be taken from density functional
⁵¹ perturbation theory (DFPT) or computed from induced dipole moments via a finite-difference
⁵² approach. RR intensities are obtained by post-processing time-dependent (Berry-phase) dipole
⁵³ trajectories obtained from RT-TDDFT simulations. Although not shown in the panel, Vibrant
⁵⁴ also supports the computation of absorption spectra from RT-TDDFT dipole data.

⁵⁵ The third panel (bottom-left) displays Vibrant's ability to dissect MD-based spectra via
⁵⁶ computing the subspectra for user-defined molecular fragments. This feature facilitates the
⁵⁷ analysis of the spectra, and is especially useful for characterizing guest-host systems, evaluating
⁵⁸ solvent contributions and molecular materials (Bas et al., 2024; Guo et al., 2010; Sun et al.,
⁵⁹ 2020).

⁶⁰ The last panel (bottom-right) demonstrates the use of Padé approximants in absorption or
⁶¹ RR spectra calculations, implemented in Vibrant through integrating the GreenX library's
⁶² analytic continuation component (Leucke et al., 2025). In RT-TDDFT simulations, frequency
⁶³ resolution strongly depends on the trajectory length, and shorter trajectories often struggle
⁶⁴ with poor spectral resolution (Bruner et al., 2016). Applying Padé interpolation is particularly
⁶⁵ useful for achieving spectral convergence from short RT-TDDFT trajectories (Bruner et al.,
⁶⁶ 2016; Mattiat & Luber, 2018). As demonstrated in the bottom-right panel, Padé interpolation
⁶⁷ enables convergence and improved frequency resolution even for relatively short trajectories,
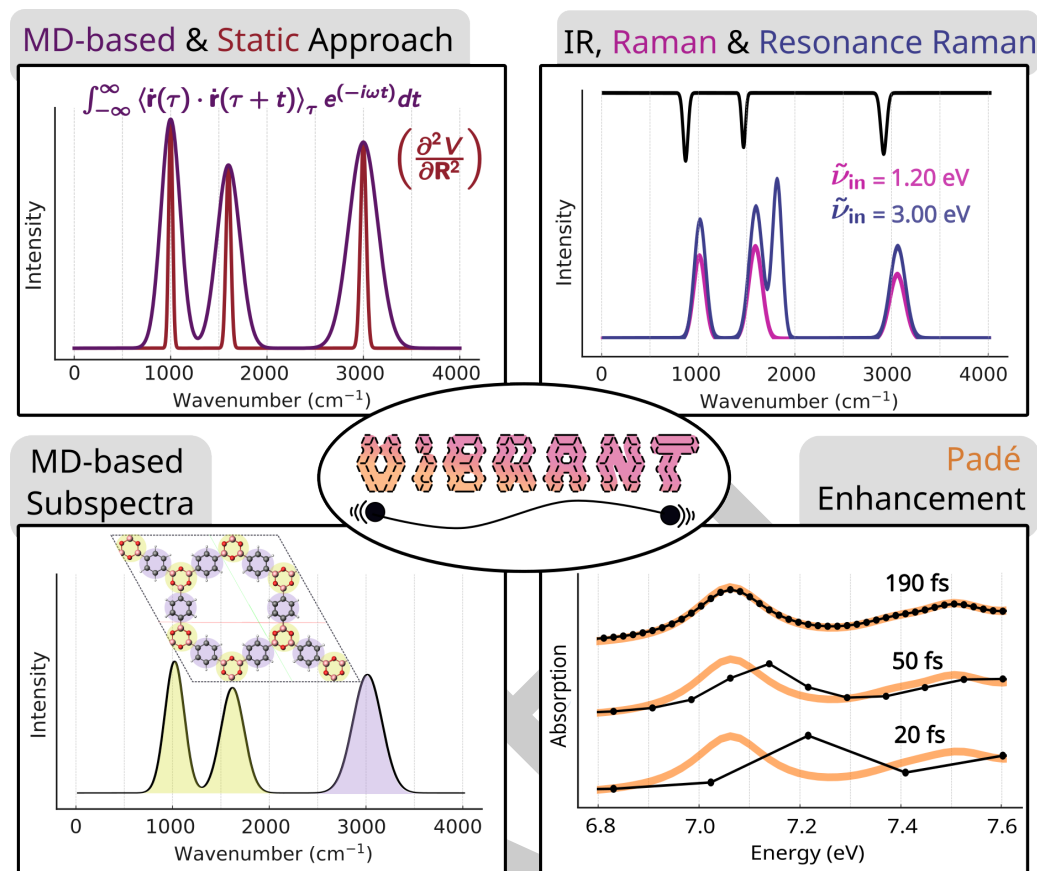⁶⁸ resulting in a fivefold reduction of overall computational cost.

Figure 1: Overview of Vibrant's functionalities. The top-left panel illustrates its ability to perform both static and MD-based vibrational spectroscopy calculations, where the vibrational frequencies are calculated from the position autocorrelation functions or the energy derivatives along displaced coordinates. The top-right panel summarizes the available corresponding spectral intensities. The bottom-left panel highlights Vibrant's feature of calculating subspectra for user-defined molecular fragments. The bottom-right panel demonstrates its integration with Padé approximants to achieve finer frequency resolution from time-dependent dipole data. More information about the theoretical background and calculation procedures are available on the Vibrant website.

State of the field

A limited number of computational programs are available for calculating MD-based and static vibrational spectra. A prominent example is TRAVIS (M. Brehm et al., 2020; Martin Brehm & Kirchner, 2011), which processes various properties obtained from MD simulations to compute different types of vibrational spectra, although it is mainly designed for liquids. There are also Python-based tools for vibrational analysis, including the scripts that process CP2K (Kühne et al., 2020) polarizabilities (Mattiat & Luber, 2018) to generate static Raman spectra (Hubmann, 2020), the scripts distributed with FHI-aims (Abbott et al., 2025; Blum et al., 2009) for normal mode analysis and static IR and Raman calculations, and VibIR-Parallel-Compute, which focuses on efficient static IR spectra (Rojas et al., 2025). For materials, a popular tool is Phonopy (Togo et al., 2023; Togo, 2023), which calculates phonon dispersion relations and vibrational densities of state; however, it is not designed for MD-based spectral analysis. Related tools include FHI-vibes (Knoop et al., 2020), which integrates Phonopy to automate vibrational and MD workflows for FHI-aims, and THeSeuSS (Boziki et al., 2025), which computes static IR and Raman spectra interfacing with FHI-aims.

To the best of our knowledge, no existing computational tool provides the same level of flexibility

as Vibrant, which supports a wide range of vibrational spectra, both static and MD-based methodologies, diverse dipole and polarizability types, and applicability across gaseous, liquid, and solid-state systems. In this context, extending existing tools such as TRAVIS was not feasible due to their limited support for community-driven development, while other mentioned packages are either much smaller in scope or not designed for broad extensibility. The need to interface with external libraries such as GreenX for computationally demanding tasks further motivated the development of Vibrant as an independent package.

Software design

Vibrant is an open-source post-processing tool for vibrational spectroscopy distributed under the Apache 2.0 license, and is designed to support both static and MD-based spectral calculations within a single and practical framework. The routines in the code are implemented in Fortran 2008 to improve performance and achieve maximized efficiency. These routines integrate established numerical libraries such as FFTW and GreenX (GX-AC), as well as OpenMP parallelization to enable efficient calculations for large systems. In addition, Vibrant is complemented by Python-based regression testing and systematic Github integration to enhance its usability and facilitate its further development.

Research impact statement

Over the course of its development, we used Vibrant in several publications to calculate and analyze vibrational spectra. The most prominent example is its application to the MD-based and static IR and Raman spectra of the layered framework material COF-1, processing different types of dipole moments and polarizabilities (Bas et al., 2024). In the same study, we also used Vibrant to investigate solvent contributions by decomposing the MD-based spectra into contributions from the framework building blocks and solvent molecules, thereby facilitating spectral analysis. In addition, we used Vibrant to generate static Raman spectra for nitrile-containing molecular C_2N frameworks (Feuerstein et al., 2025) and to compute Padé-interpolated RT-TDDFT-based absorption spectra for gas-phase naphthalene as part of the GreenX library documentation (Leucke et al., 2025). In all these studies, the results were compared against other available computational tools when applicable and showed excellent agreement with them and with experiment.

Apart from its documented applications, Vibrant provides extensive documentation covering its functionalities, implementation details, and underlying theory on its website. Provided tutorials and repositories containing input/output files of test calculations (Winogradow et al., 2025) enable user-friendly testing, while the open-source nature of the code and its regression testing support future development and extension of its features.

AI usage disclosure

No generative AI tools were used for developing the Vibrant package and preparing its documentation, authoring this manuscript, or producing the supplementary materials.

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