Molecular dynamics for modeling vibrational circular dichroism spectra and vibrational strong coupling

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Summary:
With molecular dynamics (MD), Newton's second law of motion is integrated to obtain a trajectory of positions and velocities. From this trajectory, different structural and dynamical properties of molecular systems can be obtained. One typical example is given by the vibrational properties of the system. Here we will show two recently developed examples

In a first step, MD was used to study vibrational circular dichroism (VCD) which is the weak difference in absorption for chiral molecules between right- and left- polarized light in the infrared range. VCD has promising applications in pharmacology owing to its ability to determine absolute configurations of chiral molecules. The shape of VCD spectra is highly sensitive to minor changes in conformation and molecular interactions, which makes it a sensitive probe of conformational isomerism and solvation [1].
As an alternative to methods based on explicit descriptions of electronic structure [2] and to circumvent their limited sampling capabilities in time and space, we have attempted to simulate the VCD spectrum directly from molecular dynamics trajectories employing a polarisable force field, extending earlier efforts dedicated to the IR spectrum [3]. In this presentation, we describe our implementation of VCD spectroscopy using the AMOEBA polarisable force field [4] in the Tinker software package [5].

In a second step, MD is used to explore vibrational strong coupling (VSC), which is achieved when a molecular system interacts with a cavity and forms a hybrid light-matter state. (Fig. 1) Recent experimental work has shown it is possible to reach strong coupling regimes with optical cavities [6] and that chemical reactivity is influenced [7].
The goal is to model and understand the effect of VSC on dynamics and reactivity of real molecules in solution. Thus following the work of Li et al. [8], the effect of the cavity on the properties and infrared spectrum of a periodic box of solvent molecules has been examined.

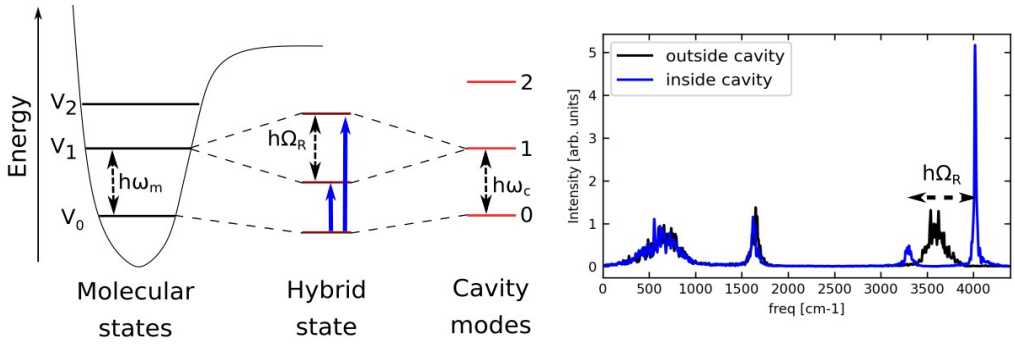

Fig. 1 Schematic representation of the coupling between a vibrational transition and an optical cavity mode (left) and the IR spectra of water influenced by a cavity mode at 3550 cm^{-1}.

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