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Title:	Understanding the Nature of Vibrational Strong Coupling in Small Molecules.
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Abstract:	<p>Strong light-matter interaction resulting in the formation of hybrid light-matter states (polaritonic states) is one of the fastest-growing research areas. It has been demonstrated experimentally that the physical and chemical properties of molecules and materials can be altered by strong coupling. For example, chemical reactivity, supramolecular self-assembly, conductivity, superconductivity etc., are modified at room temperature. However, the exact mechanism of the process is still not clear. In order to comprehend the cause of these coupling-induced modifications, it is therefore, crucial to explore thoroughly the behavior of newly generated polaritonic states. The current thesis aims to investigate the spectroscopic characteristics of these newly formed polaritonic states of small molecules in various phases. Firstly, an inhomogeneously broad OH/OD vibrational state of water/heavy water is studied systematically using advanced infrared spectroscopic tool along with transfer matrix simulations. Water vibrational states can effectively generate polaritonic states as low as 5% (v/v), and the photonic fraction persists even after detuning to a large range, indicating the versatile use of the system for polaritonic chemistry experiments. Further, linear molecules like carbon disulfide with very high oscillator strength is studied. Our experimental and analytical modeling suggest that the self-dipolar interaction term is enhanced drastically in ultra-strong coupling conditions. i.e., the actual absorption strength is boosted while coupling the oscillator to the vacuum field. The coupling strength is purely dependent on the orientation of the molecular dipoles with the electric field. Here, we used three small strained cyclic ketones (cyclobutanone, cyclopentanone, and cyclohexanone) and probed the temperature-dependent vibrational strong coupling experiments. Finally, gas phase molecules were also tested as they have the least molecule-molecule interactions. Ro-vibrational states of the gaseous nitrous oxide molecules are carried out to probe the interaction between the P- and R-branches of the molecules with the infrared cavity photon. The above spectroscopic findings and other experimental developments in polaritonic chemistry will be discussed in detail. The thesis concludes with a remark on the future perspective of polaritonic chemistry as a new spectroscopic tool to understand complex chemical reaction landscapes.</p>
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