

Time-resolved properties of vibrational polariton model

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Vibrational polariton can occur due to the coupling of an IR-active molecular transition with a resonant mode of an optical cavity. The formation of polaritons creates opportunities for new chemical reactivities by trying to manipulate the basic characteristic of photons and provide a new pathway to enable chemical transformation. This system's 2D Hamiltonian is composed of the Hamiltonian of the molecule (phonon), the Hamiltonian of the field (photon) and the Hamiltonian of the field-to-molecule (photon-to-photon) coupling and it is explored by time-independent and dynamics approaches. In a time-independent approach, one explores the eigenstates of this Hamiltonian. In a dynamic approach, one explores time-resolved propagation of the initial coherent state via the evolution operator technique. The time-dependent wavefunction is used to monitor trajectories of expectation values of several important observables. The photon-to-phonon coupling results in three major computed effects as follows: (i) It facilitates the formation of polaritons and Rabi splitting between energies of the first eigenstate and second eigenstate. The Rabi splitting has been observed to be directly proportional to the coupling strength in the small coupling regime. (ii) It facilitates change in the equilibrium bond length in the molecule and equilibrium strength of the electric field. (iii) It is responsible for the time-dependent transition of energy from molecular oscillation to field oscillation and back. The observed effects have been rationalized in terms of the coupled quantized harmonic oscillators model,¹ to quantify how the coupling strength affects the expectation value and eigenstate of the hybrid photon-to-phonon polaritonic state and also to explore the expectation values of the amplitude of the electric field and the interatomic bond displacement in the molecule. The finding of this exploration has a triple potential impact on basic science and technology: (a) Energy conversion from molecular bond oscillations to electromagnetic field amplitude serves as the foundation for the design of chemical lasers. (b) Energy conversion from cavity field to molecular motion contributes to the novel field of polaritonic chemistry for the control of chemical reactions. (c) A focus on a phase of pure quantum states is tangential to quantum information processing.

“The parameters of the model are chosen to represent ground state potential energy surface of diatomic hydrogen iodine molecule in a cavity resonantly tuned to the vibrational frequency of this molecule”

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(1) Cohen-Tannoudji, C.; Diu, B.; Laloë, F. *Quantum Mechanics*; Wiley, 1977.