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Spectroscopy and dynamics of anharmonic vibrational polaritons in the strong and ultrastrong coupling regime

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Controlling the spectroscopy and chemical reactions through molecular polaritons is one of the most studied research topics currently. However, a general theory to describe the effects that arise in the strong and ultrastrong light-matter coupling regime has not been developed yet. We model the light-matter interaction via the multi-level quantum Rabi (MLQR) model, which considers the molecular and photonic modes as anharmonic and harmonic, respectively. We show that the polariton spectrum is determined by the dipolar nature [1], which agrees with experimental measurements of a two-dimensional infrared spectroscopy of a hybrid system formed by coupling the NO band of sodium nitroprusside to a Fabry-Perot cavity [2]. We also study the dynamics of a single-hydrogen fluoride (HF) molecule coupled to a confined infrared vacuum initially prepared in the vibrational ground state, that in the ultrastrong coupling regime can be self-dissociated. We obtain dissociation probabilities of up to 20 % for conditions where additional external sources are not present [3]. Our work provides new mechanisms for possible applications in quantum metrology, quantum information processing and quantum state preparation.

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