Enhanced optical nonlinearities through strong light-matter coupling

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Abstract

Optical microcavities and metallic nanostructures have been shown to significantly modulate the dynamics and spectroscopic response of molecular systems. We present a study of the nonlinear optics of a model consisting of N anharmonic multilevel systems (e.g., Morse oscillators) undergoing collective strong coupling with a resonant infrared microcavity. We find that, under experimentally accessible conditions, molecular systems in microcavities may have nonlinear phenomena significantly intensified due to the high quality of polariton resonances and the enhanced microcavity electromagnetic energy density relative to free space. Particularly large enhancement of multiphoton absorption happens when multipolariton states are resonant with bare molecule multiphoton transitions. In particular, our model predicts two-photon absorption cross section enhancements by several orders of magnitude relative to free space when the Rabi splitting Ω_R is approximately equal to the molecular anharmonic shift 2Δ. Our results provide rough upper bounds to resonant nonlinear response enhancement factors as relaxation to dark states is treated phenomenologically. Notably, ensembles of two-level systems undergoing strong coupling with a cavity (described by the Tavis-Cummings model) show no such optical nonlinearity enhancements, highlighting the rich phenomenology afforded by multilevel anharmonic systems. Similar conclusions are expected to hold for excitonic systems that share features with our model (e.g., collections of molecular dyes with accessible $S_0 \to S_1 \to S_2$ transitions) and strongly interact with a UV-visible cavity.

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