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On the interplay of fluorescence, dissipation, and molecular dissociation for a model diatomic molecule in a quantum optical cavity.

We study the time-resolved optical response of a model diatomic molecule (Hubbard-like dimer) in a quantum cavity, and characterize how such response is influenced by the driving field, the electronic interactions and the nuclear motion, for both the second harmonic generation (SHG) and the resonance regimes. Our theoretical description is based on a full quantum and equal footing treatment of the pump and fluorescent fields, and of the electronic and nuclear degrees of freedom of the molecule. A quantum-classical approach is used to account for the cavity losses, by considering a bath of classical oscillators (Caldeira-Leggett model). By performing real-time dynamics calculations for a broad range of parameters, we find that when introducing the cavity photons via the driving field, a Mollow like spectrum occurs in the resonance regime, whereas in the SHG regime the corresponding peak vanishes. Our simulations further show that the electron interactions reduce the intensity of the fluorescent spectrum. We also observe that, with a coherent photon field as initial state, the hindrance of dimer dissociation due to increased nuclear mass enhances the SHG peak. Conversely, using a driving field to introduce the photons in the cavity, helps maintaining the Mollow-like features of the spectrum in the resonant regime. At the same time, cavity dissipation effects significantly reduce the intensity of the fluorescent spectra, and distort the Mollow-like features present in the resonance regime whereas in SHG regime the harmonic peak intensity remains the same but the Rayleigh (main) peak intensity decreases with time. While not pinned to a specific realistic system or experimental setup, our model comes through as highly expedient in illustrating general trends in the fluorescence response when dissipative effects, electronic correlations, molecular dissociation, cavity confinement, and duration of the pumping pulse are at interplay. Thus, it represents a generic and rather flexible template to gain preliminary insight for more refined investigations of systems of direct experimental interest.

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