Ultrafast nonlinear optical response of vibrational polaritons

Raphael F. Ribeiro,[1,*] and Joel Yuen-Zhou[1]
[1]*University of California San Diego, 8500 Gilman Dr. La Jolla, USA*
*rafloren@ucsd.edu

In strongly coupled molecular photonic materials, exotic and tunable excited-state (polariton) dynamics emerge with capabilities suitable for applications in the energy sciences, nonlinear optics and chemical dynamics control. To realize the potential of molecular polaritons in future quantum technologies, it is essential to understand the nonequilibrium dynamics and nonlinear optical response of molecular polaritons. This presentation will be focused on these topics.

In the first part of this talk, I will review intriguing experimental observations of ultrafast time-resolved pump-probe transmission under infrared strong coupling conditions [1-3], and subsequently provide a microscopic description of the observed phenomena based on our recently developed theoretical models. In particular, I will present and contrast theoretical treatments of non-equilibrium vibrational polariton optical response valid at (a) short pump-probe time delay, when the pump-induced excited-state population of the material is dominated by coherent polaritonic excitations [3], and (b) at long enough waiting times that the excited-state population of the system consists almost entirely of incoherent weakly-coupled molecular ("dark") states [1-2]. I will conclude by showing how our polariton pump-probe theory allows quantitative characterization of cavity-assisted vibrational energy transfer measured by coherent two-dimensional infrared spectroscopy.

References

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