Contribution ID: **3** Type: **not specified**

Analysis of cavity-suppressed reactivity via vibrational depopulation

Monday 2 October 2023 09:40 (40 minutes)

Molecular vibrations can couple to optical cavities to create new hybrid states called polaritons. Under such conditions, one may realize modified materials processes such as photon emission, molecular energy transfer, and chemical reaction rates. In this talk, I will first discuss modeling and active control of cavity coupling to molecular vibrations. Next, I will discuss measurements of reaction-rate constants extracted from evolving cavity transmission spectra, which reveal resonant suppression of the intracavity reaction rate for alcoholysis of phenyl isocyanate with cyclohexanol. We observe up to an 80% suppression of the rate by tuning cavity modes to be resonant with the reactant isocyanate (NCO) stretch, the product carbonyl (CO) stretch, and cooperative reactant-solvent modes (CH). Cavity tuning was used to selectively couple to reactant, product, and cooperative solvent-reactant vibrational modes resulting in a chemical response that is cavity tuning dependent. These results are explained using an open quantum system model that predicts resonant modifications of chemical reactivity via light-matter quantum coherences that depopulate vibrational excited states, suggesting fundamental links between chemistry and quantum science to be explored. I will show numerical experiments we carried out in search of artifacts that could impact the measurements described above. Finally, I will be briefly present the design of an experimental apparatus aimed at making liquid phase cavity-modified chemistry experiments more reliable, repeatable, and accessible for new researchers in the field.

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Session Classification: Morning session