THEORY OF INTRACAVITY NEAR-RESONANT TWO-PHOTON ABSORPTION

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I recently published a paper[1] that analyzed near-resonant two-photon absorption (TPA) spectra of polyatomic molecules excited in the vibrational fundamentals. For each molecule, a very small number of TPA lines are greatly enhanced due to near resonance. It is well known that TPA in a standing wave is dominated by a Doppler Free absorption.

In this talk, the theory will be summarised and extensions to the published paper will be presented, including: (1) Theory of high finesse optical cavities with TPA loss, including the peak transmission and spectral shape of cavity modes (which are not Lorenztian with TPA).[2] (2) Extension of the theory of saturation that includes the consequences of slow vibrational relaxation, which invalidates the traditional optical Bloch equations. (3) Theory of the TPA lineshape in the limit of low pressure such that the transition width is limited by the finite time of flight of the absorbers through the TEM_{00} mode of the cavity.[3]

1. K.K. Lehmann, J. Chem. Phys. **151**, 144201 (2019) 2. K.K. Lehmann, J. Opt. Soc. Am. A **37**, 3055 (2020). 3. K.K. Lehmann, to be published in J. Chem. Phys.