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Dynamics of Vibration-Cavity Polaritons.[1] JEFFREY OWRUTSKY, ANDREA GRAFTON, ADAM DUNKELBERGER, BLAKE SIMPKINS, United States Naval Research Laboratory — We recently reported time resolved IR pump-probe studies on strongly-coupled vibration-cavity polaritons for tungsten hexacarbonyl in hexane in a Fabry-Pérot cavity [1]. While much of the response is due to reservoir or uncoupled excited state absorption as well as polariton contraction, a component of the observed signals is due to polariton state evolution. We subsequently used low concentration samples, which reduce the Rabi splitting between the polaritons, to highlight the polariton contraction and demonstrate saturable absorption not only in pump probe measurements but also in single pulse studies.[2] We further investigated this system with two-dimensional infrared (2D IR) spectroscopy which provides evidence of hybrid light-matter polariton evolution and clear indications of direct excitation of dark states.[3] We have further expanded the investigation of vibrational dynamics for strongly coupled vibration-cavity polaritons to another solute, nitroprusside in methanol We explore salient features of the transient response, especially at short delay times, which show aspects of the response that are due to polaritons and are distinguished from uncoupled higher order excitations. [1] A. D. Dunkelberger, et al., Nat. Comm. 7, 13504 (2016). [2] ACS Photonics 6. 2719 (2019). [3] B. Xiang, R. F. Ribeiro, A. D. Dunkelberger, J. Wang, Y. Li, B.S. Simpkins, J.C. Owrutsky, J. Yuen-Zhou, W. Xiong, PNAS 115, 4845-4850 (2018).

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